

SYNTHETIC FUELS: A PERSPECTIVE ON COMMERCIALIZATION

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Introduction

The commercial production of liquid and gaseous fuels derived from coal and oil shale is currently hindered by a variety of complex and interacting factors. Conversion processes are highly capital-intensive, and their products are at best estimated to be only marginally competitive with landed world crude oil and imported LNG at current prices. Added to this is a multitude of constraining factors, such as resource availability, protection of the environment, sociopolitical considerations, marketplace logistics, financing, the potential effects of competing new technology, and the possibility of wider availability of international supplies of lower-cost crude oil and natural gas.

Strongly countering this situation is a growing realization of the long-term need for supplemental supplies of gaseous and liquid hydrocarbons as we enter an era in which depletion of conventional resources is a worldwide concern. Although recovery and conversion will be costly, the United States has substantial reserves of coal and oil shale that can provide supplemental fuels. This situation, together with a national goal of reducing dependence on oil and gas imports, will lead to increasing pressure for the commercialization of synthetic fuels.

This paper presents summary comparative economics estimated for synthetic fuels from coal and oil shale and focuses on some of the important financial considerations associated with the commercialization of synthetic fuels. Some of the pertinent conclusions are:

- Upgraded shale oil liquids are estimated to be less expensive than coal-derived liquids and can be marginally competitive with imported crude oil.
- Based on recent field pilot work, speculative economics look attractive for low-Btu gas by underground gasification of coal. Large-scale demonstration appears desirable.
- A financing mechanism sheltered from excessive risk will probably be necessary to encourage commercial synthetic fuels ventures.

The summary economics presented are derived from a variety of studies recently performed at SRI involving the comparative evaluation of synthetic fuels from coal and oil shale.

Status of Technology

Considerable research and development activity in the United States has recently been devoted to the conversion of solid fossil fuels into the more desirable energy forms--clean gases and liquids. This is far from a new concept. Coal has been gasified to produce town gas, synthesis gas, or reducing gas on a worldwide scale since early in the 19th century. Underground coal gasification (UCG) has been commercially practiced in the USSR since around 1940. Coal liquids have been recovered from the pyrolytic coking operations associated with steel manufacturing since early in the industrial revolution. More recently, during World War II, conversion of coal to liquid fuels reached a production level of approximately 100,000 barrels per day in Germany; and coal liquefaction is currently practiced commercially on large

scale in South Africa. However, most of this recent production of liquids was, and continues to be, by Fischer-Tropsch synthesis from coal-derived gases ($\text{CO} + \text{H}_2$). In Estonia, recovery of liquids from oil shale began commercially in the mid-1930s. Since that time, developmental scale oil shale retorts have been operated in several countries including the United States, Brazil, and China.

Two notable coal gasification systems that are actively marketed throughout the world and commercialized in several countries are the familiar Lurgi fixed-bed gasifier and the Koppers-Totzek entrained gasifier. Other gasification systems marketed commercially are: Winkler, McDowell-Wellman, Woodhall-Duckham, Riley-Morgan, and Wilputte.

Coal conversion systems at or nearing large-scale development in the United States or with direct U.S. involvement are listed in Table 1. Also shown are oil shale retorting systems that have been developed on a large scale in the United States and have been indicated for use in potential commercial ventures. Of the coal conversion systems listed, the Lurgi gasifier, as previously mentioned, has already been widely commercialized for synthesis gas and town gas. It is mentioned here as a developing system because of its potential extension to production of SNG or fuel gas, in modified form to permit gasification of caking coals, and for close coupling with electric power generating systems.

In addition to the developmental oil shale retorting systems listed in Table 1, large-scale projects are also currently active in Brazil (Petrosix process) and in Estonia (the Soviet-developed Galoter process). Plans have been announced (1), (2) in those countries to build commercial-scale plants using the two processes.

Three of the developmental coal conversion processes mentioned--BG-Lurgi slagging, COGAS, and Coalcon--have been chosen by the U.S. Energy Research and Development Administration (ERDA) at this time for more detailed study to determine suitability for ERDA/industry jointly sponsored large commercial-scale demonstration. Evaluation of the first two systems for SNG continues while the future of the Coalcon "Clean Boiler Fuel" demonstration program is now in considerable doubt. In addition, ERDA and a variety of industrial organizations are supporting the thrust toward demonstration of low- or intermediate-BTU gas (LBG/IBG) systems with the "Combined Cycle Test Facility" and ERDA's "Gasifiers in Industry Program," which is aimed at the small and medium sized user, and their "Hydrogen from Coal Program."

Economic Analysis

Cost is normally the overriding consideration in bringing a new technology into the marketplace. In this regard, the most visible competition in the United States for gaseous and liquid fuels from coal and oil shale (unless there are significant new finds of oil and gas in North America) is imported world crude oil which is currently landed at \$13 to \$15 per barrel (\$2.20 to \$2.50 per million Btu) and imported LNG which will be somewhat more expensive. Additional competition for the use of synthetic fuels includes direct firing of coal for power generation or industrial heat (with or without flue gas desulfurization), and eventually natural gas transported from northern Alaska or the Canadian frontier.

Most of the conversion processes listed in Table 1 have been technically and economically evaluated by SRI in the course of a variety of recent project work. Summary process cost ranges are presented in Table 2. Capital costs are presented in terms of dollars of plant investment per daily production output in barrels of oil equivalent. Product costs are compared both on the basis of U.S. regulated utility economics (cost of service/rate base) and the discounted cash flow (DCF) rate of return analysis commonly used for commercial ventures. Resulting annual capital charge rates are 29 to 32 percent for the 15 percent DCF rate of return cases and 15 to 17 percent for the regulated utility analysis. All costs are in terms of mid-1977 constant dollars.

With the highly debt-leveraged and relatively financially sheltered regulated utility analysis, estimated process costs vary from \$1.50 per million Btu for the speculative economics associated with low-Btu gas from underground coal gasification to \$5.00 per million Btu for methanol from coal by near-term commercial technology. These costs increase to \$2.00 and \$7.25 per million, respectively, for a 100 percent equity DCF analysis yielding a 15 percent rate of return. On the same DCF basis, costs for synthetic crude oil from oil shale are in the range of \$2.75 to \$3.50 per million Btu (\$16 to \$21/barrel) for developing retorting and upgrading systems. The regulated utility analysis results in costs 25 to 30 percent lower than those for the DCF analysis. Product costs shown in Table 2 are compared and discussed in more detail later in the paper.

A few caveats concerning the development of these costs are clearly in order. The coal costs specified are intended to represent a mine-mouth conversion plant location and therefore do not include coal transportation charges. Furthermore, the costs presented in Table 2 are battery limits process costs and do not include specific site-related charges that can be significant, depending on the remoteness of location. Some of these potentially important site-related factors are: water development projects (including pipeline and storage); mine to conversion-plant railroads; power transmission lines; access roads; product pipelines; extensive environmental analysis; and townsite development activities.

These, together with other potential site-specific requirements, could easily increase capital investment by 10 to 20 percent and could add \$2.00 to \$3.00 per barrel (\$0.35 to \$0.50 per million Btu) or more to product cost for a synthetic fuels venture in a remote location. Product cost ranges, based on Table 2, but including provision for such site-specific costs, are compared in Figure 1. Excessive delay in attaining design capacity and plant modifications during start-up of new technology are other factors that will add significantly to costs for first-of-a-kind plants. Over the life of the project, these items could easily contribute additional product costs in the range of \$1.00 to \$3.00 per barrel (\$0.20 to \$0.50 per million Btu). The costs summarized in Figure 1 do not include contingency for pioneer plants.

There is some hope of process "learning curve" cost reduction over near-term technology with the evolutionary development of advanced technologies. Development of processes that eliminate or combine process steps could potentially lead to significantly lower product costs. An example in the case of SNG could be a system successfully combining process steps such as gasification, shift conversion, and perhaps reducing the gas cleanup and methanation requirements, or eliminating the need for an oxygen plant. Such processes will undoubtedly contribute additional costs of their own, but significant potential exists for lower overall costs. In the case of LBG/IBG, lower costs could potentially result from successful development of a gasifier using a sulfur acceptor or combined with some other type of hot gas cleanup system. The lower cost ranges in Table 2 and Figure 1 represent such advanced or innovative processes. Without such development, it is doubtful that cost reductions of greater than about 10 percent will be possible with foreseeable conversion technology, since such systems will probably be composed primarily of plant sections based on already commercial or near-commercial technology.

Outlook for Commercialization

Clearly, the considerations concerning commercialization will vary somewhat for SNG from coal, LBG/IBG from coal, coal liquids, and oil shale. In view of the status of technology, the economics of production, and other factors, what are the prospects for commercialization? The following prognosis is offered.

SNG from Coal: The estimated cost of SNG from western U.S. subbituminous coal using conventional Lurgi gasification technology in a coalfield location is estimated to be \$3.75 to \$4.25 per million Btu (coal at \$7 per ton), which is clearly not competitive directly with natural gas marketed either on an interstate or intrastate basis. However, a massive and intricate distribution system exists for transmitting energy in the form of high-Btu gas directly to the consumer. Therefore, while interfuel competition at the marketplace will be a prime consideration, there will be strong incentive to use these existing distribution systems for SNG. Potential direct alternative competitors for SNG from coal in the United States are imported LNG and natural gas transmitted from the northern frontier areas of North America. However, these alternatives will also involve extremely large front-end investment, and some will have the additional disadvantages associated with security of supply. Of course, energy conversion, transmission, and distribution via electrification is a competitor, but again the costs for that alternative are also significant (3), and comparative costs are highly dependent on location.

Some SNG technology is now near commercial and additional systems are ready for commercial-scale demonstration. However, the cost of conversion plants will often be large in relation to the capitalization of companies planning to build such plants, thus making financing difficult without some type of guarantee for the lending institution. If regulatory approval necessary for the gas pricing structure permits rolling-in of the higher cost of SNG, then commercialization would probably follow, although it is likely that some type of debt guarantee will also be required.

Because of necessary regulatory approvals, the probable need for federal financial sheltering, and the lead-time necessary for plant construction, it is doubtful that any plants based on Lurgi technology could be on stream before 1985. However, with regulatory and financial approaches now under discussion, some commercial operation appears likely in the period 1985-1990.

LBG/IBG from Coal: LBG/IBG from bituminous coal in an eastern U.S. location without site-related factors included is estimated to cost about \$3.25 per million Btu for commercial technology and ranging to about \$2.50 per million Btu for some developmental processes.

High conversion plant investment and production costs will limit markets for LBG/IBG in the near term; however, some markets may develop in the 1980s due to the curtailment of natural gas, or where the direct use of coal or heavy oil is excluded. Potential markets for LBG/IBG technology include utility power generation (4), industrial fuel (5), synthesis gas as a chemical feedstock, and reducing gases for the primary metal industries. The production of synthesis gas from coal may be most attractive in areas of the United States where chemical manufacturing is heavily concentrated, therefore permitting construction of large central conversion facilities.

The logistics of coal and gas are important when comparing competitive markets for SNG from coal with LBG/IBG from coal. For example, coal supply, distribution, and handling considerations may add significantly to the cost for the small LBG/IBG producer. Therefore, an SNG-from-coal facility feeding existing natural gas distribution networks may deliver high-Btu gas to local markets competitively with small-scale systems producing LBG or IBG for those markets. Moreover, even when large centralized production facilities are used, both LBG and IBG will require limited but expensive gas distribution systems which may make delivered SNG less expensive. This type of comparison is highly site-specific, and generalization is difficult.

Recent experimental field pilot work (6) conducted by ERDA's Laramie Energy Research Center (LERC) in the Hanna Basin in Wyoming indicates progress toward development of an operable underground coal gasification system in

the United States. Speculative economic estimates indicate process costs for low-Btu gas of about \$1.50 per million Btu on the basis of regulated utility economics. Remote site-related costs could increase this to about \$2.00 per million Btu. Coalfield gas-fired power generation will be the primary market for this gas. Recent cost estimates (7) indicate that this may be competitive with mine-mouth power generation by direct firing of coal with flue gas desulfurization. Based on recent LERC experience, it appears that developmental costs for UCG are in some cases probably the lowest of any of the synthetic fuel types. UCG will eliminate most of the objections of stripmining of coal and can permit utilization of thick deep or otherwise difficult to mine coal seams, although surface subsidence and disruption and contamination of aquifers are potentially serious problems. Underground gasification control problems will have to be identified and overcome before low-Btu gas from UCG can be used in its most logical market--coalfield-located power generation. UCG technology is in the early stages of development in the United States; however, based on current field test results, large-scale demonstration now appears desirable to verify speculative economics, prove necessary operability, and assess environmental effects. Future commercialization will, of course, be dependent on demonstration results. Development is not far enough advanced at this time to predict when commercial application might occur.

Coal Liquefaction: Coal-derived liquids appear to face a more difficult competitive situation than coal-derived gases, since they will compete directly with imported oil in petroleum liquids markets. Liquids from oil shale will also compete directly in these markets.

Costs for low-sulfur liquids and boiler fuel with developmental systems are estimated here to be in the range of \$3.50 to \$4.50 per million Btu (\$21 to \$26 per barrel) for a 15 percent DCF rate of return. The regulated utility financing analysis does lower this to the \$15 to \$19 per barrel range. However, site-specific factors could easily add \$2 to \$3 per barrel to the costs.

Production of methanol from coal by gasification and catalytic methanol synthesis is somewhat more expensive than direct liquefaction of coal because of lower conversion efficiencies (40 to 50 percent versus 65 to 70 percent) and significantly higher capital costs. Based on a DCF rate of return analysis, the cost of methanol from coal is estimated at a little over \$7.00 per million Btu using near term commercial technology before providing for remote site costs. Even assuming cost reductions due to evolutionary improvements in technology, and when using a highly debt-leveraged regulated utility economic analysis, the estimated cost of methanol from coal exceeds \$4.00 per million Btu or \$22.00 per oil-equivalent barrel on the process basis used. DCF costs on the same basis are approximately \$6.00 per million Btu or about \$35 per oil-equivalent barrel.

Large pilot plants for the direct production of liquids from coal are now in varying stages of design, construction, and operation. However, because of the high estimated costs, it is doubtful that there will be any significant commercial production of liquids from coal, other than from potential government-sponsored commercial scale demonstration programs, before the mid-1990s, at the earliest. Furthermore, there appears to be little prospect of methanol being produced from coal in the United States in the foreseeable future.

Oil Shale: Even though this paper is part of a session addressing the comparative economics of coal conversion processes, it is appropriate that production of liquids from oil shale be considered.

Based on a variety of SRI studies, the costs of an upgraded synthetic crude oil using developmental retorting systems are estimated (Table 2) to be in the range of \$2.75 to \$3.50 per million Btu (\$16 to \$21 per barrel) for a

discounted cash flow rate of return of 15 percent. The equivalent costs for regulated utility financing are in the \$12 to \$15 per barrel range. If it is feasible to produce and ship raw shale oil or a lightly upgraded high nitrogen content product, the cost would be \$3 to \$5 per barrel less at the plant than for a highly hydrotreated, low-nitrogen synthetic crude. Even though site-specific factors could easily add \$2 to \$3 per barrel to the above costs, shale syncrude with a sheltered financial basis appears to be marginally competitive with imported oil (Figure 1).

Shale oil product costs are estimated to be significantly lower than costs for coal-derived liquids. However, it should be emphasized that the greatest technical and economic risks in oil shale production are in shale mining and disposal of spent shale. In the case of coal liquefaction, uncertainties associated with the mining of coal and disposal of ash are considerably less. Large-scale pilot testing of several retorting technologies has been carried out. Some of this technology is now ready for commercial-scale demonstration. Such a demonstration in addition to proving retorting technology reliability would provide answers to serious questions concerning mining technology, overall economics, and environmental effects.

The lead-time necessary for construction of a commercial-scale plant would now probably preclude oil shale operation prior to about 1985, even with an early decision to construct a plant. In general, with access to world oil prices and a sheltered financial position, together with resolution of environmental constraints, some commercial production should develop in the period 1985-1990.

Any discussion at this time of the commercialization of synthetic fuels is not complete without addressing the issue of potential federal government incentives. During the 1973-1976 period, at least three industrial groups were willing to initiate commercial SNG-from-coal projects, assuming their SNG pricing structure was approved by the Federal Power Commission. No additional federal incentives or financial sheltering were apparently felt to be necessary to assist commercialization at that time. Moreover, at least four industrial-sponsored oil shale projects were set to move forward without federal assistance; indeed, the oil shale groups in most cases paid large front-end federal lease bonuses for the "opportunity" to do so.

Cost escalation has since magnified the risk of these ventures to a level where the sponsors have indicated that some type of financial incentive and/or risk reduction mechanism is now necessary. Four potential incentives often discussed to assist the commercialization of synthetic fuels processes are:

- Federally guaranteed construction loans
- Product price supports
- Tax credits
- Construction grants.

These incentives are discussed in detail in the report summarizing results of the Synfuels Interagency Task Force Study in 1975 (8). Since 1975, Congress has focused legislative attention on the use of loan guarantees. While this technique is now being sought by regulated producers for construction of SNG plants, there is no such consensus about loan guarantees among nonregulated producers for ventures involving synthetic liquids plants.

An alternative to the incentive approach is a modification of the so-called government-owned, company operated (GOCO) approach. During World War II, processes for the production of high octane gasoline and synthetic rubber were developed with federal assistance. The federal government provided the risk capital for initial construction and operation of pioneer commercial-scale demonstration plants and

when the plants were commercially proven, they were sold on a bid basis to the industrial developer, or to some other company. This type of approach may now be necessary to assist construction of pioneer plants, particularly for coal liquefaction, oil shale, and second-generation SNG systems now under development.

Summary and Overview

Commercial viability of synthetic fuels operation in the near term is clearly questionable because of the competition with natural gas and imported oil. Based on the economic analyses discussed in this paper, LBG/IBG from coal and liquids from oil shale appear to have the best chances for near-term commercial application. However, financial success appears to be contingent on economics based on a sheltered financial position to permit rates of return characteristic of a regulated utility producer. SNG from coal, while estimated to be more costly than LBG/IBG and shale liquids, may see earlier commercialization because of an existing sheltered regulatory position and because existing transmission, distribution, and utilization systems are available for the product.

Estimated costs for developmental conversion processes may be reduced by perhaps 20 percent or a little more with evolutionary technology, but greater cost reductions are difficult to foresee. A notable exception is underground coal gasification which, while being extremely site-specific and at an early stage of development in the United States, can have attractive economics.

Incentives currently discussed may be sufficient to commercialize first-generation coal gasification systems; however, an alternative such as government-financed plant construction and operation prior to successful commercial demonstration with sale to the developer/operator may be necessary for commercialization of pioneer technologies. Even when analyzed using the most attractive financial situation, the costs of synthetic fuels (UCG is a potential exception) will be at best marginal with to well above the current and near-term projected prices of natural gas and imported oil.

It becomes increasingly apparent that it is desirable for the United States to expand its ability to develop additional indigenous supplies of hydrocarbons, both to increase the base of a secure supply and to reduce the potentially adverse effect that excessive imports will have on the U.S. trade balance. However, the free market driving force for near-term large-scale commercialization of solid fossil fuel conversion processes is tenuous. Some specific marketing situations will develop; but in the near- to intermediate-term these will be caused more by shortages of conventional fuels rather than by price competition. In the meantime, large-scale research and development of a variety of systems has reached a significant level of sophistication and accomplishment. Commercial-scale demonstration of selected technologies because of the long lead-time necessary to commercialize such technology is now desirable to assist in the selection of the best near- to intermediate-term processes and for the benefit of long-term ongoing research and development.

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Table 1

MAJOR COAL CONVERSION AND OIL SHALE RETORTING SYSTEMS NOW UNDER DEVELOPMENT

COAL GASIFICATION

<u>Name</u>	<u>Type</u>
• SNG	
- Lurgi	Fixed-bed, dry-ash
- BG-Lurgi	Fixed-bed, slagging
- HYGAS	Fluid-bed
- CO ₂ Acceptor	Fluid-bed
- BIGAS	Entrained
- Synthane	Fluid-bed
• LBG/IBG ¹	
- Lurgi	Fixed-bed, dry-ash
- BG-Lurgi	Fixed-bed, slagging
- Texaco (Partial Oxidation) ²	Entrained
- Combustion Engineering	Entrained
- Battelle (Agglomerating Ash)	Fluid-bed
- Westinghouse	Fluid-bed
- GEGAS	Fixed-bed
- Underground gasification	Laramie ERC technique

COAL LIQUEFACTION

• Solid Boiler Fuel	
- Solvent Refined Coal	Noncatalytic
• Low-Sulfur Oil	
- H-Coal	Direct catalytic
- Exxon Donor Solvent	Indirect catalytic
- Synthoil	Direct catalytic
- Coalcon	Noncatalytic

OIL SHALE RETORTING

• Liquids Production	
- TOSCO II	Circulating Heat Carrier
- Paraho	Internal Combustion
- Occidental	In situ

¹Low or intermediate Btu gas

²This general type of technology is also under development jointly by the Royal Dutch Shell Group and Krupp/Koppers

Table 2: ESTIMATED CAPITAL AND PRODUCT COST FOR SOLID FUEL CONVERSION SYSTEMS

(1) (Battery Limit Basis)				(7) Product Cost			
Coal Gasification	Coal Type	Battery Limit Process Capital Investment	MS/Daily O.E. Bbl (2)	Regulated Utility Analysis		Discounted Cash Flow Analysis	
				\$/MMBtu	\$/O.E. Bbl	\$/MMBtu	\$/O.E. Bbl
SNG	Lurgi	(3)	24	3.75	22.00	5.25	30.75
	Developmental Processes		18-24	2.75-3.75	16.00-22.00	4.00-5.25	23.50-30.75
LHG/IRG	Lurgi	(4)	19	3.25	19.00	4.75	26.25
	Developmental Processes		15-21	2.50-3.50	14.50-20.50	3.50-5.00	20.50-29.25
Coal Liquefaction (6)	Underground Coal Gasification		8	1.50	8.75	2.00	11.75
	Sub. Bit.						
Oil Shale (6)	Low-Sulfur Liquids and Boiler Fuel		13-15	2.50-3.25	15.00-19.00	3.50-4.50	21.00-26.25
	Methanol		21-25	3.75-5.00	22.00-29.25	5.50-7.25	32.25-42.50
Synthetic Crude Oil (5)	-----		12-15	2.00-2.50	11.50-14.50	2.75-3.50	16.50-20.50

NOTES:

- (1) Battery limits costs, site-specific factors are excluded
- (2) Oil equivalent barrel (O.E. Bbl): 5.85 million Btu
- (3) Powder River Basin
- (4) Illinois No. 6
- (5) Piceance Basin
- (6) Developmental Processes, low-sulfur/nitrogen products
- (7) Plant gate product costs

BASIS:

- Mid-1977 constant dollar analysis
- Regulated Utility Analysis: 65% debt, 15% return on equity, 10% interest on debt
- Discounted Cash Flow Analysis: 15% DCF rate of return, 100% equity financing
- Plant size: SNG : 250 x 10⁶ scf/day (1,000 MW equivalent)
LHG/IRG : 5 x 10⁹ Btu/hour (500 MW equivalent)
Liquids : 100,000 O.E. Bbl/day (2,500 MW equivalent)
- Mine Mouth Coal Prices: Powder River Basin Subbituminous = \$7/ton
Illinois No. 6 Bituminous = \$18/ton
- Site-specific costs for a remote location may add \$2 to \$3 per O.E. Bbl (\$0.35 to \$0.50 per million Btu).

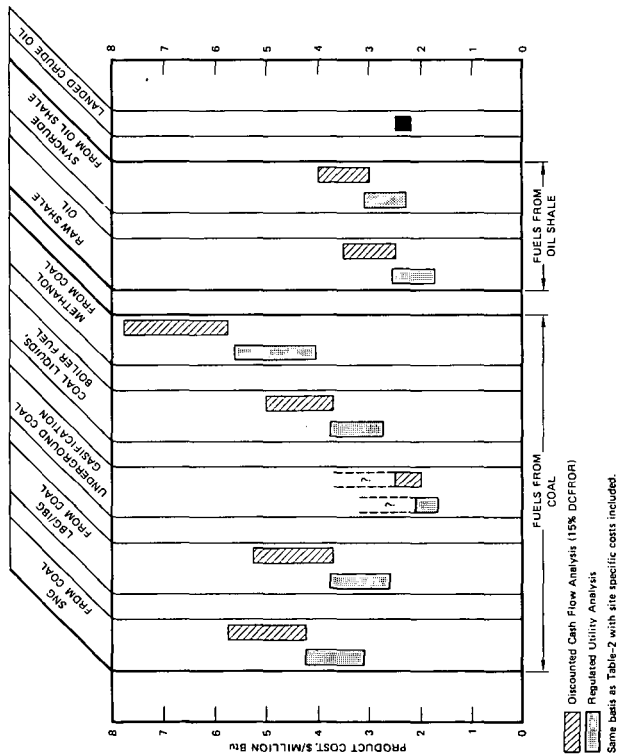


FIGURE 1 ESTIMATED PLANT GATE COST FOR SYNTHETIC FUELS FROM COAL AND OIL SHALE (Mid-1977 Constant Dollar Analysis)

Same basis as Table-2 with site specific costs included.

Coal Conversion Plant Cost Escalation

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Coal conversion plants are capital intensive. At least half the price of coal-derived synthetic fuels arise from fixed charges (1) and the cost of commercial-sized plants are such that capital availability is a major problem. Thus, economic evaluations of coal conversion processes are strongly influenced by the capital cost estimates used. Further, these estimates in recent years have been subject to substantial escalation due to inflation. Also, the long periods of planning and construction required for coal conversion plants compounds both the effects of escalation and interest during construction.

Since escalation heavily impacts coal conversion economics, a study was undertaken for ERDA to: 1. Identify and quantify factors contributing to the recent escalation of coal-based fuel plant cost estimates. 2. Identify indices which in the past have best reflected inflation escalation of coal conversion plants, and 3. Suggest procedures for predicting future capital cost escalation.

Escalation analysis requires at least two cost estimates of the same plant made at different times. Preferably there should be several sets of estimates for several plants all using the same process and having about the same capacity. The only available sets of estimates meeting the above requirements were cost estimates for the Lurgi coal gasification process. These estimates were particularly valuable since they were based on common design data supplied by the Lurgi Corp. In addition, the three sets of estimates used were associated with plants which all used Western sub-bituminous coal to produce from 250 to 288 million SCFD of pipeline grade gas per stream day.

Prior to the main analysis, the applicability of Lurgi results to other gasification processes was checked by comparing Lurgi escalation data with estimated escalation for several advanced gasification processes under comparable conditions. These data derived from (2) and (3) are tabulated below.

<u>Process</u>	<u>Aug. '74 Cost,</u> <u>\$MM</u>	<u>Jan. '76 Cost,</u> <u>\$MM</u>	<u>% Increase</u>
IGT Hygas	750	870	16
CO ₂ Acceptor	760	890	17
BCR Bi-Gas	890	1,030	16
Lurgi	920	1,060	15

From the above, it appears that Lurgi escalation data can be extrapolated to advanced pipeline grade coal gasification processes.

Low and medium Btu gas projects present a special problem since each plant is

designed to meet a particular customer's demand. (Typically, capacities vary from 21 to 100 trillion Btu [year].) However, since the reactor and purification steps of the low Btu processes are similar to those of the Lurgi processes, there appears to be no a priori reason why escalation for pipeline grade gas should be markedly different from escalation for lower grade gas processes.

Coal liquefaction presented another problem. Aside from methanol (around 85 percent of the Lurgi and methanol processes capital costs are for common equipment), liquefaction and gasification processes do not have a high degree of equipment commonality. Consequently, the approach taken was to perform the analysis outlined below and then check the main results with experts in coal liquefaction technology. In brief, the results for inflation escalation did check fairly well.

Lurgi cost estimates were collected and then normalized to place them on a comparable basis as possible. The normalized data were then disaggregated into the components of escalation that arise from: 1. Changes in the type of estimate. 2. Changes in knowledge of technology and environmental requirements. 3. Changes in equipment capacity, and 4. Changes in the costs of equipment, materials and labor (inflation escalation).

The inflation component of escalation was then compared by statistical tests and other means with various cost indices. Then, based on certain criteria, those indices best reflecting estimated Lurgi escalation were identified. The results and implications were analyzed and, finally, methods for predicting future escalation were suggested.

Five organizations generously cooperated in the collection and analysis of the estimates. These were, El Paso Natural Gas Co., Panhandle Eastern Pipeline Co., Southern California Gas Co., The Fluor Corp. and the Lummus Corp. To protect the privacy of the estimates, results are identified only as venture A, B or C.

The component of escalation due to estimate type was first examined. This component could arise because a prudent venture manager might require that a contingency adjustment equal to the upper bound of the accuracy range of the pertinent type of estimate be included in project costs. However, none of the estimates obtained had to be adjusted for estimate type because all of the estimates were "preliminary" (about $\pm 20\%$ probable accuracy) and all contingency costs were removed when the estimates were normalized.

After removal of contingency, interest during construction and certain project specific costs (e. g., coal mine and sales tax costs) the following breakout of escalation for the ventures was obtained.

Lurgi Escalation Results

Venture	Escalation						Comments
	Sept. '72-July '73		July '73-Jan. '75		Jan. '75-Early '76		
	Tech. %	Inflation %	Tech. %	Inflation %	Tech. %	Inflation %	
A	38	6	34	56	-	-	Tech. escalation did not include design.
B			38	57	8	20 (Jan.)	Tech. escalation includes design changes.
C			N. A.	60	N. A.	16 (Apr.)	Estimates adjusted to remove technology and design change escalation.

Note that estimates were in constant dollars as of estimate date, i. e., no forward escalation.

There are four indices that are commonly used to track inflation escalation. These are the Chemical Engineering (CE), Marshall & Swift (M&S), the Nelson index of refinery costs (Nelson) and the Engineering News Record (ENR) indices. Also, coal-fired power generation boiler plant equipment escalation as given by the privately circulated Handy & Whitman (H&W) index was thought pertinent.

The five indices identified above are based on weighted averages of labor, material and equipment costs. But it was suspected that the traditional weighted-average indices understate the importance of equipment prices during periods of high inflation such as those experienced between mid '73 and early '75. Consequently, two pure equipment indices were also compared to the Lurgi data. These were the Nelson equipment index (a composite of five equipment classes) and an index of valve and fitting prices which was the only single item index that matched Lurgi escalation reasonably well. The results of the comparison are tabulated below.

Comparison of Lurgi and Cost Indices Inflation

Escalation

Category	Escalation		
	Sept. '72-July '73	July '73-Jan. '75	Jan. '75-Early '76
	%	%	%
Lurgi	6 (1)*	56-60 (3)*	16-20 (2)*
<u>Chemical Engineering</u>			
(CE)	5	24	4
Marshall & Swift			
(M&S)	2.5	28	5
Nelson Refineries	5	19	7
<u>Engr. News Record</u>			
(ENR)	8.5	11	9
Handy & Whitman			
(H&W)	6	37	9
Nelson Equipment	4	38	8
BLS Valves & Fittings	5	51	6

* Numbers in parentheses indicate number of observations.

Since the Lurgi estimates only covered about four years, it was thought desirable to select a process which might exhibit about the same inflation escalation as a Lurgi process and where data was sufficient to permit development of an index for a number of years. The ethylene cracking process was selected for the following reasons: 1. Both Lurgi and ethylene plants are reasonably similar from the standpoint of scaling factors. 2. The front end of a Lurgi plant includes a number of identical gasifiers; the front end of an ethylene plant includes a number of identical cracking furnaces. 3. Both types of plants have either single or dual purification systems and substantive compression equipment. Also, they require closely integrated utility systems. 4. Escalation for the ethylene index for 1973-1975 was 67 percent vs. 55-60 percent for Lurgi from mid-1973 to 1975.

The processes are different in that gasification plants have provision for extensive solid handling facilities for coal feed and ash removal. But, for other reasons, coal preparation and handling costs were backed out of the estimate during the normalization procedure.

The ethylene index was used as follows. The coefficient of correlation "r" between the rates of change of the ethylene (Lurgi analog) index and the other indices were computed. Rates of change were used to reduce the autocorrelation between successive values of an index. The "r" values were then used as one criterion for selection of a representative index. The other criterion was closeness of fit to gasification escalation during the critical inflation period of mid '73 to early '75. The evaluation results are shown below.

Index Evaluation Results

Index	Escalation from July '73-Jan. '75 %	Correlation Relative to Ethylene	
		1965-1975 "R"*	1970-1975 "R"***
Lurgi Plants	56-60	N. A.	N. A.
Ethylene Plants	N. A.	1.000	1.000
CE	24	0.828	0.758
M&S	28	0.736	0.636
ENR	11	-0.134	-0.754
Nelson	19	0.491	0.152
Nelson Equipment	38	0.814	0.759
H&W	37	0.735	0.627
Valves	51	0.715	0.708

*R > | .625 | Significant at > .95 Level

**R > | .811 | Significant at > .95 Level

The results are also illustrated in Figures 1 and 2. Figure 1 shows the rates of change of three representative indices and the ethylene index, while Figure 2 shows the actual ethylene values vs. values predicted from the three indices. The predicted values were obtained by first regressing ethylene values on the indicated indices to obtain linear equations with relative ethylene costs as the dependent variables. Then, actual values of the appropriate indices were inserted into the equations to generate the prediction curves in Figure 2. All predicted values underestimate inflation in 1974, and then overcompensate in 1975.

The results obtained for the Lurgi process were then reviewed with experts in coal liquefaction. What was learned is summarized below.

	<u>Lurgi/Ethylene Data</u>	<u>Coal Liquefaction Data</u>
Technology	85% for Lurgi, Sept. '72	15% over 4 years
Escalation	to Jan. '75	
Inflation	195% Mid '70-Mid '75, C ₂ H ₄	250% Mid '70-Mid '75
Escalation	55-60% Mid '73-Early '75, Lurgi	Lurgi escalation "in the ball park" for liquefaction.

It was concluded that the high Lurgi process technology escalation is not typical of other coal conversion processes in that: 1. A World War II period design had to be adapted to modern materials and fabrication techniques. 2. The downstream design (purification, shift and methanation) had to be adapted to larger capacities than originally specified. 3. The design had to be adapted to U.S. codes and environmental regulations. The implication of this conclusion is that aggregated cost data such as that presented in Lurgi ventures to the FPC may

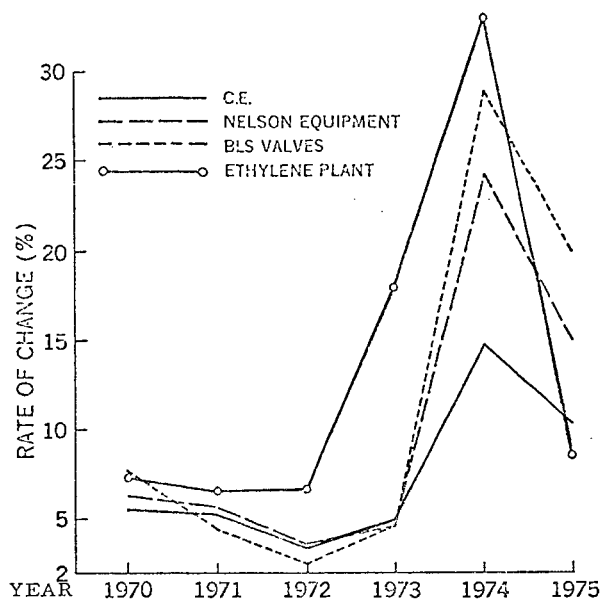


Figure 1. RATE OF CHANGE VS. YEAR

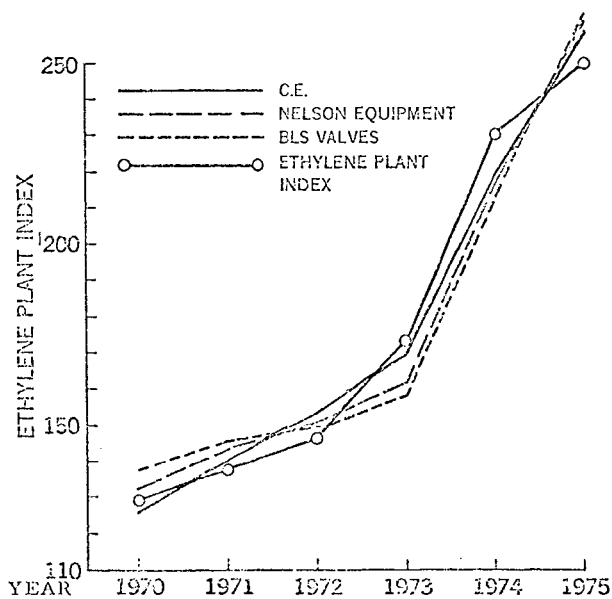


Figure 2. PREDICTED INDEX VALUES VS. YEAR

exaggerate the amount of escalation that might be experienced by advanced coal conversion processes.

On the other hand, it was concluded that the inflation escalation estimated for the Lurgi process was generally applicable to other coal-based synthetic fuel processes. Using the criterion of fit to high inflation periods, the index for valves was judged to best reflect the inflation escalation experienced in the critical 1973-1974 period. Using the correlation coefficient criterion, either the CE or Nelson equipment index was judged "best".

It was also concluded that an equipment type index tracks escalation during periods of high inflation better than do the weighted-average indices. (Note the CE index is weighted 61 percent for equipments vs. 12 percent for the Nelson index vs. 0 percent for the ENR index.)

Methods suggested for predicting future escalation included the following:

- Correlate certain representative indices (e.g., as tentatively identified above) with exogeneous indicators by econometric methods. Forecasts of the indicators are available from such services as DRI, Chase and Wharton.
- Survey the larger constructor/engineers for 2-4 year trend forecasts on equipment, material and labor costs pertinent to coal conversion plants.
- Survey a cross-section of process equipment manufacturers to obtain detailed cost trends.

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ECONOMICS OF COAL GASIFICATION PROCESSES FOR ELECTRIC POWER GENERATION. PACK, G. E.
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92715

Economic aspects of large coal gasification-power generating plants are presented. These aspects include thermal efficiencies, capital cost and cost-of-services. Existing and advanced gasification technologies are investigated. Several concepts that have been proposed are explored.

CONVERSION OF COAL TO LIQUIDS BY
FISCHER-TROPSCH AND OIL/GAS TECHNOLOGIES

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INTRODUCTION

Conversion of coal to liquid and gaseous fuels as well as chemical products has been practiced on a commercial scale in several areas of the world. Projections of U.S. supply and demand balances for crude oil and natural gas to the year 2,000 indicate that coal conversion plants are a candidate in the U.S. for production of environmentally acceptable liquid and gaseous fuels. To be competitive with alternative energy sources, second generation production complexes for coal conversion should be large, efficient, simple and reliable.

This paper describes the characteristics and projected economics for two candidate second-generation technologies, "Oil/Gas" and a U. S. version of Fischer-Tropsch.

The term "Oil/Gas" was coined during the 1974 Project Independence Blueprint campaign. The process uses a type of coal hydroliquefaction similar to SRC II, with reaction severity designed to produce a significant amount of light hydrocarbons. These are in turn processed to yield substitute natural gas (SNG) as a prime product. Liquid products include LPG, naphtha, and fuel oil.

The suggested U.S. version of Fischer-Tropsch incorporates flame-sprayed catalyst on extended heat-exchanger surfaces yielding several potential advantages including increased thermal efficiency. Flame-sprayed catalyst systems have been under development by what is now the Pittsburgh Energy Research Center (PERC) for about 15 years.

The information presented here is based primarily on conceptual designs and economic evaluations prepared by The Ralph M. Parsons Company for the Major Facilities Project Management Division of Energy Research and Development Administration - Fossil Energy (ERDA-FE).^{1,2} The conceptual design given for each process incorporates certain process and equipment items now under development, primarily within ERDA programs. The designs are intended to define the potentials for second generation coal conversion complexes incorporating results of in-progress development work. In concept, these complexes might be constructed and operated in the mid-'80's to mid-'90's.

This paper will describe the processing, projected product characteristics, and projected economics for the Fischer-Tropsch and Oil/Gas complexes. These factors will then be compared, recognizing that each produces significantly different products. In addition, the Oil/Gas design¹ will be extended by hydrotreating fuel oil to produce lower percent sulfur products at increased cost, to further illustrate the flexibility of the technology.

Each of these conceptual designs represents only one of numerous possible configurations. For a given industrial application with a defined coal source and required product mix, the design would in actual practice be tailor-made for that particular case.

OIL/GAS

Design Criteria

Preliminary design criteria have been published.³ Key elements of the completed conceptual design are:

<u>Plant Location</u>	Eastern region of the U.S. Interior Coal Province, which includes portions of the states of Illinois, Indiana, and Kentucky.
<u>Coal Source</u>	Illinois No. 6 seam coal produced in a captive surface coal mine.
<u>Capacity</u>	Approximately 47,000 tons per day (TPD) of run-of-mine (ROM) coal which is cleaned, washed and sized to produce about 36,000 TPD of coal feed to the process plants. All daily figures are in stream days. Products include about 165 million standard cubic feet per day (MM SCFD) of SNG and approximately 75,000 barrels per day (BPD) of liquids consisting of LPG, naphtha, and fuel oil.
<u>Plant Availability</u>	The plant is considered to operate at capacity 330 stream days per year, resulting in an availability factor of 90.4%.
<u>Characteristics</u>	The complex is a grass roots facility which cap- tively produces all utilities and oxygen require- ments. All effluent streams are treated to meet environmental standards.
<u>Raw Material and Product Storage</u>	Facilities are provided for a 14-day coal inventory and a 30-day liquid product inventory.

Facility Description

An artist's conceptual drawing is presented in Figure 1. A photograph of a model of the complex is shown in Figure 2. The complex would occupy approximately 600 acres, exclusive of the coal mine. Plant population is about 2,350 people. About 17,500 gallons of water per minute would be drawn from the source river.

<u>Coal Mine</u>	The mine is an integrated strip mine with five separate areas or mining units to produce 47,000 TPD of ROM coal operating 350 days per year. The average overburden is 60 feet and average coal seam thickness is 5 feet. The primary overburden removal is with 170 cubic yard drag-lines. The ROM coal would pass through a primary separation step located in the mining area
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and then be transferred by conveyor to a coal preparation plant area where it is cleaned and sized to produce feed coal to the process plant.

Over the 20-year project operating life, approximately 57 square miles would be mined out.

Process Plant

A process block flow diagram is shown in Figure 3.

Key to the process is the SRC II hydroliquefaction step. Here, 20,000 TPD of cleaned, sized feed coal is slurried in coal-derived recycle solvent; two-thirds of the solvent is unfiltered and contains undissolved coal and ash, while the remaining one-third has been filtered to remove the solids. The coal slurry is pumped to 2,050 psig, mixed with hydrogen, preheated to 700°F, and reacted in the dissolver vessel. The dissolver product passes through a pressure let-down system with the resulting liquid phase going to a low pressure fractionator. Fractionation products are naphtha, light distillate used as fuel oil constituent, heavy distillate used as filter wash oil and as a product fuel oil constituent, and the bottoms which contain solids. The bottoms are split; about half are recycled to the feed coal slurry system and the remainder goes to the filters.

The naphtha is hydrotreated to produce saleable product. The light distillate, a portion of the heavy distillate, and the filtrate are combined to form the product fuel oil.

Gases emitting from the dissolver pressure let-down system, fractionation, and the naphtha hydrogenation steps are combined and fed to a monoethanolamine (MEA) acid gas removal system to take out the hydrogen sulfide, carbon dioxide and carbonyl sulfide. The resulting sweet product gas is then processed in a cryogenic unit for hydrocarbon recovery/separation as described below. Sour acid gas is sent to a sulfur plant which removes the sulfur-containing contaminants and produces saleable sulfur.

In the SNG and LPG production train, sweet gas produced in the MEA system is dried with molecular sieves and then sent to a cryogenic unit. Here 98.5 volume percent hydrogen is recovered. A portion of this hydrogen stream is used to methanate residual carbon monoxide. Then the high purity hydrogen is fed to the naphtha hydrotreater while the remainder of the hydrogen stream is recycled to the coal dissolving step. Methane-rich gas produced in the cryogenic unit is compressed, cooled to remove condensable fractions, and then passed through a zinc oxide guard chamber to reduce the hydrogen sulfide content. It is then processed in a final methanation unit and sent to the SNG product line. Ethane and heavier fractions produced in the cryogenic unit are fractionated to remove ethane and some propane overhead which is mixed with final methanator product to produce specification grade SNG, which is compressed to 1,000 psig for delivery. Remaining propane and heavier material is separated into propane LPG as an overhead product and a bottoms product. Bottoms are debutanized to produce butane LPG as an overhead product, leaving pentane-and-heavier bottoms which are fed to the naphtha hydrogenation unit. Butane LPG is hydrotreated

and transferred to product storage.

The make-up hydrogen stream for the coal dissolving step is produced in a coal-fed gasifier operated at about 1,000 psig. Significant methane is produced at this pressure. The gasifier is a two-stage entrained slagging type. Solids are removed from the gasifier effluent gas stream and the hydrogen-to-carbon monoxide ratio adjusted in a sour shift conversion unit. The shifted gas is processed in a physical solvent acid gas removal system to produce a sweet gas for feed to the dissolver section, a hydrogen sulfide-rich gas stream for feed to the sulfur plant, and a carbon dioxide-rich vent gas stream. The Rectisol process was used as a representative process.

A fuel gas gasifier system is included to generate the necessary steam and power to operate the complex. This gasifier is fed by the dried dissolver filter cake plus coal. The filter cake is previously dried to recover the wash solvent as a saleable product. Fuel gas generated in the gasifier is treated in an acid gas removal system to remove hydrogen sulfide and carbon dioxide before passing to the power and steam generation section.

Power and Steam Generation

The in-plant produced fuel gas is used to produce electrical power in two condensing turbine generator units with three extraction points. Four steam boilers are also included. The utility system is closely integrated with the process plant operation.

Material Balance

The overall material balance for the process plant is shown in Figure 4. Material inputs consist of coal, water, and oxygen (from the air separation plant). The coal amounts to about 36,000 TPD. Saleable products, including fuels, sulfur and ammonia, add to approximately 19,000 TPD.

Energy Balance

The energy balance is depicted in Figure 5. The projected thermal efficiency, coal to saleable products, is about 77%.

FISCHER-TROPSCH

Design Criteria

Preliminary design criteria have been described.⁴ Key elements of the completed conceptual design are:

<u>Plant Location</u>	Eastern Region of the U.S. Interior Coal Province.
<u>Coal Source</u>	Illinois No. 6 seam coal produced in a captive surface coal mine.
<u>Capacity</u>	Approximately 40,000 TPD of ROM coal will be mined and 30,000 TPD of cleaned, sized coal will be fed to the process plant. The products will have an energy value of approximately 525 billion Btu per day consisting of 260 MMSCFD of SNG and approximately 50,000 BPD of liquid products which are LPG's, light and heavy naphthas, dissel fuel, fuel oil and oxygenates.

<u>Plant Availability</u>	350 stream days per year; availability factor = 90.4%.
<u>Characteristics</u>	Grass roots facility producing all utilities plus oxygen and treating all effluent streams to meet environmental standards.
<u>Raw Material and Products Storage</u>	Fourteen-day coal storage and 30day liquid product storage.

Facility Description

The complex is depicted in the artist's conceptual drawing shown in Figure 6 and a photograph of model of the complex is presented in Figure 7. Land area required for the complex, without the coal mine, is about 500 acres. Plant population is about 2,100 people. Approximately 12,000 gallons per minute (GPM) of water would be required.

Coal Mine

As in the Oil/Gas design, a strip mine with an average overburden of 60 feet and average seam thickness of 5 feet would produce the required 40,000 TPD of ROM coal. The mine would consist of four integrated mining faces. The primary separation and coal preparation units are similar to those previously described for the Oil/Gas complex with the exception that the ground coal has a smaller particle size; minus 20 mesh by 0 for Fischer-Tropsch vis-a-vis 5% plus 20 mesh, 25% minus 200 mesh for the Oil/Gas plant.

Process Plant

All of the feed coal is fed to two entrained slagging-type steam oxygen gasifiers operated at approximately 470 psig. Gasifier effluent gas stream is exhaustively cleaned to remove solid particles. The ratio of hydrogen to carbon monoxide in the cleaned gas is increased by subjecting about 50% of the gas stream to a shift conversion reaction; the H_2/CO ratio is thereby adjusted to the target value of 1.45. Shifted gas is then fed to an acid gas removal unit where it is contacted with a physical solvent to remove the hydrogen sulfide, carbon dioxide and organic sulfur compounds. The Selexol process was used as a representative process for this design. The absorbed acid gases are stripped for further processing; the hydrogen sulfide is converted to saleable sulfur in the sulfur plant and the CO_2 stream is vented. Sulfur content of the cleaned syngas is reduced to about 0.1 part per million, volume (ppmv).

Cleaned syngas is fed to the Fischer-Tropsch synthesis unit at about 400 psig and 570°F. It first passes through zinc oxide guard chambers to remove trace quantities of sulfur compounds. Then it is processed in 18 parallel synthesis reactors designed for isothermal operation. The reactors have flame-sprayed iron catalyst deposited on the external surface of extended surface heat exchangers. Reaction takes place on the shell side and 1,250 psig steam is generated on the tube side by the heat of reaction. Shift and methanation reactors have a similar geometrical design but differ in the composition of the flame-sprayed catalyst.

Fischer-Tropsch reactor feeds contain a ratio of recycle to fresh feed of approximately 1.4. Extensive heat exchange is used to maintain a high plant thermal efficiency.

Fischer-Tropsch products go to a liquid product recovery unit to recover light hydrocarbons from the Fischer-Tropsch gas and to fractionate the liquids into the product streams.

Two gas streams are recovered and fed to the methanation unit which produces SNG. One consists of a mixture of residual lean gas after absorption of the C_3 's in a presaturated lean oil stream and a CO-rich stripper overhead product produced by stripping a lean oil fractionator overhead stream. This mixed stream is fed to the first methanation stage. It contains gases produced in the Fischer-Tropsch reactor, including methane and some C_2 's and C_3 's to increase the heating value of the SNG. An additional feed stream, which goes to the second-stage methanator, consists of C_3 's and C_4 's which are produced in a depropanizer in the liquid product refining train; they serve to increase the heating value of the SNG.

Fischer-Tropsch liquids are preheated and fed to a lean oil fractionator where light ends are removed overhead for further processing and feed to the methanator section as described previously. The bottoms are fed to the fuels vacuum fractionator where the heavy naphtha, diesel oil and heavy fuel oil are produced. Naphtha is removed as an overhead product. Diesel oil is withdrawn as a side stream and is steam-stripped to obtain the flash point specification. Heavy fuel oil is produced by steam stripping in the bottom section of the fractionator, cooled, and sent to storage.

Light naphtha is produced in a naphtha stabilizer fed by the bottoms from the depropanizer. C_4 LPG's are recovered as overhead from the stabilizer.

Oxygenate produced in the Fischer-Tropsch reactor, containing a high alcohol content, are recovered and refined. Feed to the oxygenate recovery system is produced in a water extraction of the Fischer-Tropsch liquids. This feed is preheated and the oxygenates taken overhead from a fractionator with the bottoms returned to the extraction system. A hot alcohol-salt solution, produced by caustic neutralization of the Fischer-Tropsch reactor effluent to destroy acids produced in the reaction, is stripped and the oxygenates recovered as an overhead product are also fed to the oxygenate fractionator previously discussed. The stripper bottoms are evaporated to produce a concentrated salt solution for disposal and a condensate stream used as boiler feed water.

Product SNG is produced in the methanation section. The primary feed is sulfur-free stripped gas produced in the liquid product recovery section. The methanation section consists of a first-stage recycle reaction unit containing three methanators in parallel, and a second-stage one-pass finishing reactor.

Feed gas to the first stage methanator is mixed with 1.25 parts of recycle gas, preheated to about 570°F, and reacted at 380 psig in isothermal reactors of design similar to those used for the Fischer-Tropsch reaction. A flame-sprayed nickel catalyst is deposited on the outside surface of a finned tube heat exchanger and the high heat of reaction removed by boiling dowtherm in

the tubes -- the hot downthorn in turn is used to generate 1,300 psig steam for use in the plant utility system. Reaction conditions in the first-stage methanator favor CO methanation to assure that the product SNG does not contain more than 0.1 mol% CO. Product from this first-stage methanator is cooled, condensate removed, and about three-fourths of the gas recycled with the remainder going to the second-stage methanator which is an adiabatic fixed-bed radial-flow reactor using a pelleted, reduced, nickle-type catalyst. Here the CO₂ is methanated; it will also methanate CO if a breakthrough should occur in the first stage. The CO₂ content of the product SNG is maintained below 2.5%.

The product from the second-stage methanator has a higher heating value of about 910 Btu/SCF. This is combined with the vaporized mixed light hydrocarbon stream produced in the liquid product recovery section and fed to a hydrotreater for saturation of alkenes by the residual hydrogen in the stream. The product SNG stream is cooled, condensate removed, compressed, dried, and fed to the product pipeline at 1,000 psig.

POWER AND STEAM GENERATION

The process produces all steam required for operations, heating, and power generation. Therefore, conventional steam boilers are not provided for normal operation. A start-up boiler is provided.

Electrical power is generated by four 120-megawatt extraction steam turbine generators. These generators provide all power required for operation of the complex plus approximately 140 MW for sale.

MATERIAL BALANCE

Overall material balance for the process units is presented in Figure 8. Results indicate that approximately 13,000 tons per day of saleable fuel products plus sulfur are produced from 30,000 tons per day of cleaned, sized feed coal.

ENERGY BALANCE

Energy balance is summarized in Figure 9. Estimated thermal efficiency in converting coal to saleable products is approximately 70%.

PRODUCT CHARACTERISTICS

Projected product characteristics for the Fischer-Tropsch and Oil/Gas conceptual designs are summarized in Table 1. These have been projected based on review and analysis of product characteristics reported by process investigators for similar, but not identical, process conditions^{5,6}, plus minor adjustments to reported product characteristics using the characterization factor to assure consistency with the basic data. For more radical adjustments to reported product characteristics as a result of subsequent treatment, for example, hydrogenation, reference was made to published work⁷ in this area to establish change of characteristics resulting from treatment. There are not yet reports of production-analysis-functional product testing of large quantities of the naphtha, diesel fuel, and fuel oil streams. However, the projection of these characteristics based on analysis of existing data and comparison of expected values based on analogy to other coal-derived liquids

and similar crude oil-based products provides a basis for projecting comparative results for these two technologies and defining incentives for pilot plant production to permit confirmation or modification of the projections.

The most significant differences are that the Fischer-Tropsch liquid products contain nil sulfur, nitrogen and particulate matter, and are composed primarily of aliphatic compounds, while the Oil/Gas products contain sulfur, nitrogen, and solids and consist primarily of aromatics. The Fischer-Tropsch liquids therefore have higher potential for use as petrochemical feedstocks and for fuel applications with stringent environmental restrictions. Oil/Gas products show promise for use in gasoline manufacture and for selected fuel applications. Additional comments will be presented later regarding possible market values of these products.

FIXED CAPITAL INVESTMENTS

All economics are expressed in Fourth Quarter 1976 dollars.

The projected fixed capital investments (FCI) for the two conceptual complexes are compared in Table II. The results indicate that the Fischer-Tropsch complex would require a FCI of approximately 1.55 billion dollars to produce about 85,000 barrels of fuel oil equivalent per day (BOE/D); the FCI per BOE/D is therefore about \$18,000. The Oil/Gas complex would require a FCI of about \$1.3 billion to produce approximately 110,000 BOE/D for a FCI per BOE/D of about \$12,000.

A comparison of the relative costs of the separate sections of the complex is shown in Table III. A significant contributor to the higher FCI per BOE/D for the Fischer-Tropsch plant lies in the gasification section where the cost of the oxygen plants and gas cleanup are much higher. Note that the FCI's for the conversion sections, per daily barrel of oil equivalent, for the two complexes are about equal.

TOTAL CAPITAL INVESTMENTS

Total capital investments are presented in Table IV. Total capital includes fixed capital investment, initial catalyst and chemicals, start-up costs, construction financing, working capital, and land/rights of way. Projected total capital requirements are 2.0 and 1.7 billion dollars for the Fischer-Tropsch and Oil/Gas complexes, respectively. Example construction financing costs are presented in each case.

Estimated time to mechanical completion was approximately 57 months in each case. This included design, engineering, procurement and construction.

OPERATING COSTS

Projected annual operating costs for the complexes are given in Table V. The operating costs include royalty allowance of \$1.50 per ton of cleaned, sized coal produced.

Projected annual operating costs are 205 million dollars for both the Fischer-Tropsch and Oil/Gas complexes, respectively. For analytical purposes, the complexes were divided into cost centers.

REQUIRED PRODUCT SELLING PRICE

Average required product selling price was projected for three project financial structures. In all cases, the project operating life was 20 years.

- o 100% equity capital
- o Borrowed capital: 65% of the total investment borrowed at 9% interest, with the principal repaid in equal installments over a 20-year project operating term; all working capital borrowed at 9% interest for the 20-year term; a loan commitment fee of 0.75% on funds not drawn down during the construction period.
- o A nonprofit (0% discounted cash flow rate of return) or break-even boundary case.

A 12% discounted cash flow rate of return (DCF) was selected as a base case, and the revenue required to achieve this DCF calculated for each financial structure. Required average product selling price was then calculated using the required revenue and the quantity of energy products produced.

Results are summarized in Table VI. Here we see that for the 65/35 debt/equity financial structure, the projected average required product selling prices, fourth quarter 1976 basis, (RPSP) are \$2.55 and \$1.95 per million Btu's for the Fischer-Tropsch and Oil/Gas cases, respectively. The 100% equity financing cases are about 30 percent higher in each case. The breakeven cases are about \$1.50 and \$1.20 per million Btu's, respectively.

In dollars per barrel, the 65/35 debt/equity case RPSP's would be about \$15.25 and \$12.00; this is based on an arbitrary 6 million Btu per barrel reference value. A key factor in the economic projections is the inclusion of large captive coal mines in the complexes.

SENSITIVITIES

Sensitivities of the average required product selling price to changes in key economic parameters are shown in Table VII. The RPSP is most sensitive to changes in fixed capital investment. To illustrate, for Fischer-Tropsch a 10% reduction in fixed capital investment would result in an 8.7% reduction in RPSP for the 100% equity case. The sensitivities to operating costs are in the range of 15-20%.

Effect of variations in DCF on the RPSP is presented in Figure 10 for the 65% debt case. Sensitivity is greater for the 100% equity case, which is not shown.

POSSIBLE PRODUCT MARKET VALUES

A brief assessment of possible product market values and the effect of the resulting project revenues on profitability was completed. To obtain these possible market values, the project characteristics of the products were compared with those of conventional crude oil-based products. Discussions were held with representatives of fuel producers and consumers and industry reports were reviewed.

Industry representatives strongly qualified their opinions on possible prices by stating that laboratory and field product performance tests must be conducted before firm dollar values could be assigned to the products.

With the above caveats clearly in mind, possible unit sales values and annual revenues for a fourth quarter 1975 basis are presented in Table VIII for Fischer-Tropsch and Table IX for Oil/Gas; these are taken from the published reports.^{1,2} The SNG sales value was based on value allowed for sale of SNG produced commercially from naphtha, and possible values for SNG from coal at that time. These possible sales values are presented to illustrate the effect of product sales value on the economics and also to perhaps stimulate further effort to establish firm product values and marketability.

The Tables VIII and IX possible annual revenues were then updated to a fourth quarter 1976 basis using Federal Energy Administration data which indicated that fuel prices escalated approximately 9 percent from fourth quarter 1975 to fourth quarter 1976.

Results of this second-order exploratory analysis indicate that possible average annual revenues (Fourth Quarter 1976 dollars) are \$730 and \$560 million dollars for the Fischer-Tropsch and Oil/Gas cases, respectively. Projected DCF's calculated using these revenues and the project structures developed earlier are shown in Table X. To illustrate, for the 65/55 debt/equity case, the projected DCF's for Fischer-Tropsch and Oil/Gas are 27 and 20 percent, respectively. This result indicates the incentive for accurate assessment of the marketability and profitability of synfuel products to be produced in second generation coal conversion plants in the U.S.

ECONOMIC COMPARISON FOR LOW SULFUR CONTENT FUEL PRODUCTS.

Projected sulfur content of the Oil/Gas fuel oil is 0.45. A brief and very preliminary analysis of the effect of further hydrotreating to reduce the sulfur content on cost and product composition was made; this is an extension of the design previously reported.¹ The result provides guidance regarding the costs and implications of producing very low sulfur fuels from coal by Oil/Gas type technology for environmental reasons.

The data basis for predicting process and cost results for hydrotreating the coal-derived liquids is limited.^{3,10} However, some information is available to guide the projections.

Preliminary process designs were developed for incremental hydrotreating of the Oil/Gas fuel oil. Hydrotreating conditions were nominally 650°F and 2,500 psig with a nickel-molybdenum type catalyst. A 6 months catalyst life was assumed for the purpose of this preliminary assessment.

Projected product distribution as a function of fuel oil sulfur content is depicted in Figure 11. With decreasing sulfur content, the amount of fuel oil decreases and the lighter products increase.

Figure 12 presents projections of hydrogen consumptions and Figure 13 shows projected required average product selling price at 12% DCF, 65% debt as a function of fuel oil sulfur content. Also shown on Figure 13 is the projected

RPSP for nil sulfur Fischer-Tropsch products. Results indicate that at about 98% sulfur reduction in Oil/Gas fuel oil, the required product selling prices are approximately equal.

SUMMARY AND CONCLUSIONS

Conceptual designs' economic evaluations for two candidate second generation coal conversion technologies have been completed by the Ralph M. Parsons Company. These are a suggested future version of a Fischer-Tropsch complex, and an Oil/Gas Complex which uses SRC II technology. Each conceptual design incorporated certain process and equipment concepts currently under development. The designs are based on the presumption that these development programs will be successful.

The conceptual complexes process 30,000-36,000 tons per day and produce 85,000-110,000 barrels per stream day of oil equivalent. Projected fixed capital investments for the Fischer-Tropsch (F-T) and Oil/Gas (O/G) complexes are 1.55 and 1.3 billion dollars, respectively; all economics are presented in fourth quarter 1976 dollars. Unit fixed capital investments, expressed as dollars per daily barrel oil equivalent (BOE/D) are about \$18,000 and \$12,000, respectively.

Projected product characteristics from the complexes differ; Fischer-Tropsch produces primarily aliphatic liquids and Oil/Gas primarily aromatics.

Projected required selling prices to achieve a 12% DCF using a 65% debt, 9% interest case are about \$15.25 and \$12.00 per equivalent barrel. A second order assessment of possible product sales values has led to the conclusion that DCF's of the order of 20% might be achieved; this is presented to illustrate the incentive to produce and test enough of the synfuels to determine their market values.

Projections of possible costs for hydrotreating a 0.4% sulfur Oil/Gas fuel oil to reduce its sulfur content have been presented. Results indicate that reducing the sulfur content to 0.1% would add an incremental \$500 million to the fixed capital investment and reduce the quantity of fuel oil by about 6 percent while increasing the quantities of LPG's and naphtha. A further result is a 15 percent increase in the average required product selling price (RPSP). The average RPSP at this sulfur level is projected to be about 90% of the nil sulfur F-T RPSP. At 98% sulfur reduction in Oil/Gas fuel oil, the RPSP's are about equal. Limited information is available for this hydrotreating step. An incentive exists to develop a firm basis for design and prediction of economics.

Fischer-Tropsch and Oil/Gas coal conversion technologies each offer different advantages and potential problems to be overcome. They must be considered candidates for any future synfuels-from-coal programs.

ACKNOWLEDGEMENT

There are many contributors to designs/assessments of this scope. The guidance of Messrs. D. Garrett and N. P. Cochran of the Major Facilities Project Management Division of ERDA - Fossil Energy is gratefully acknowledged. Also, the contributions of Messrs. A. Bela, S. M. Fass, G. H. Hervey, R. D. Howell, H.W. Klumpe, B. I. Loran, E. A. Mills and D. G. Reynolds, all of Parsons.

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4. O'Hara, J. B., Bela, A., Jentz, N. E., and Khaderi, S. K., "Fischer-Tropsch Plant Design Criteria," Presented at the 68th Annual Meeting of the American Institute of Chemical Engineers (AIChE), Los Angeles, Calif., November 19, 1975.
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10. Ralph M. Parsons proprietary data files.

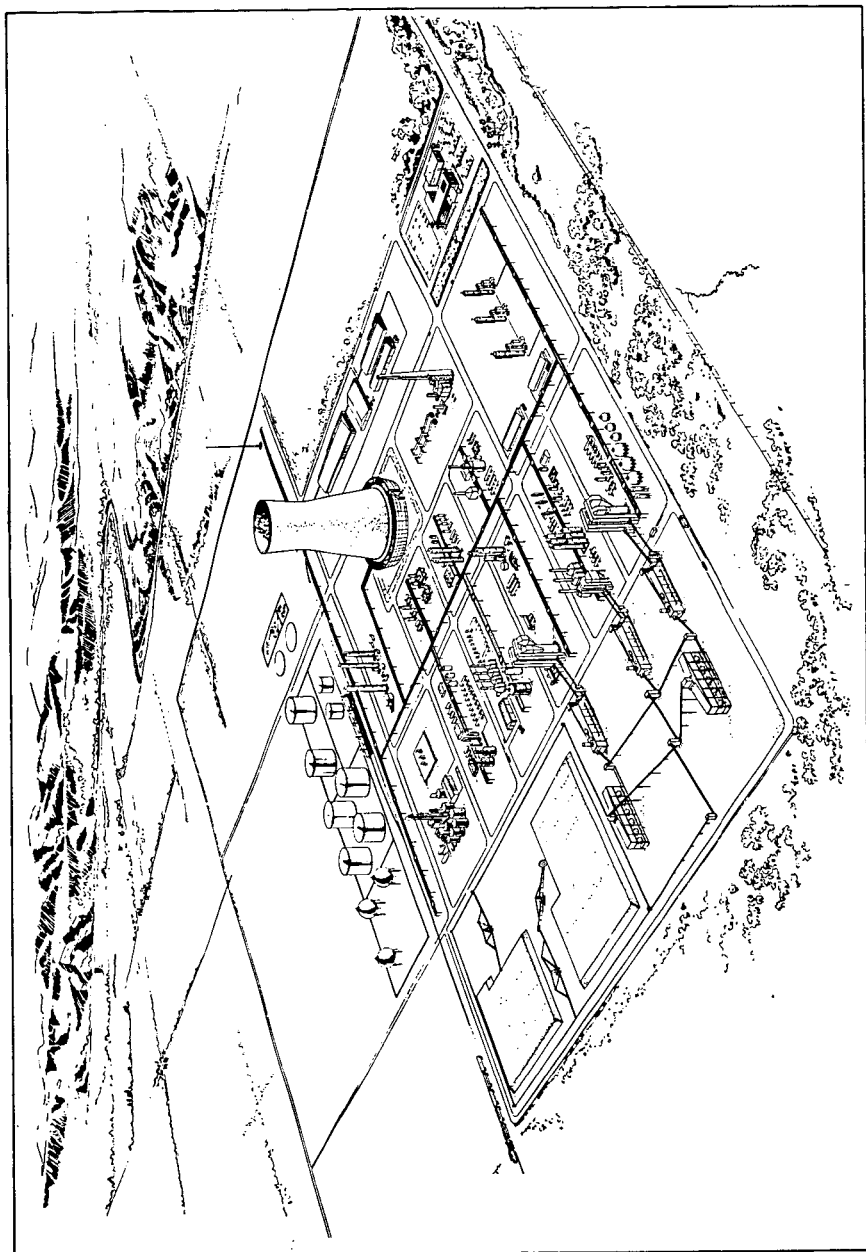


Figure 1 - Artist's Concept, Oil/Gas Plant

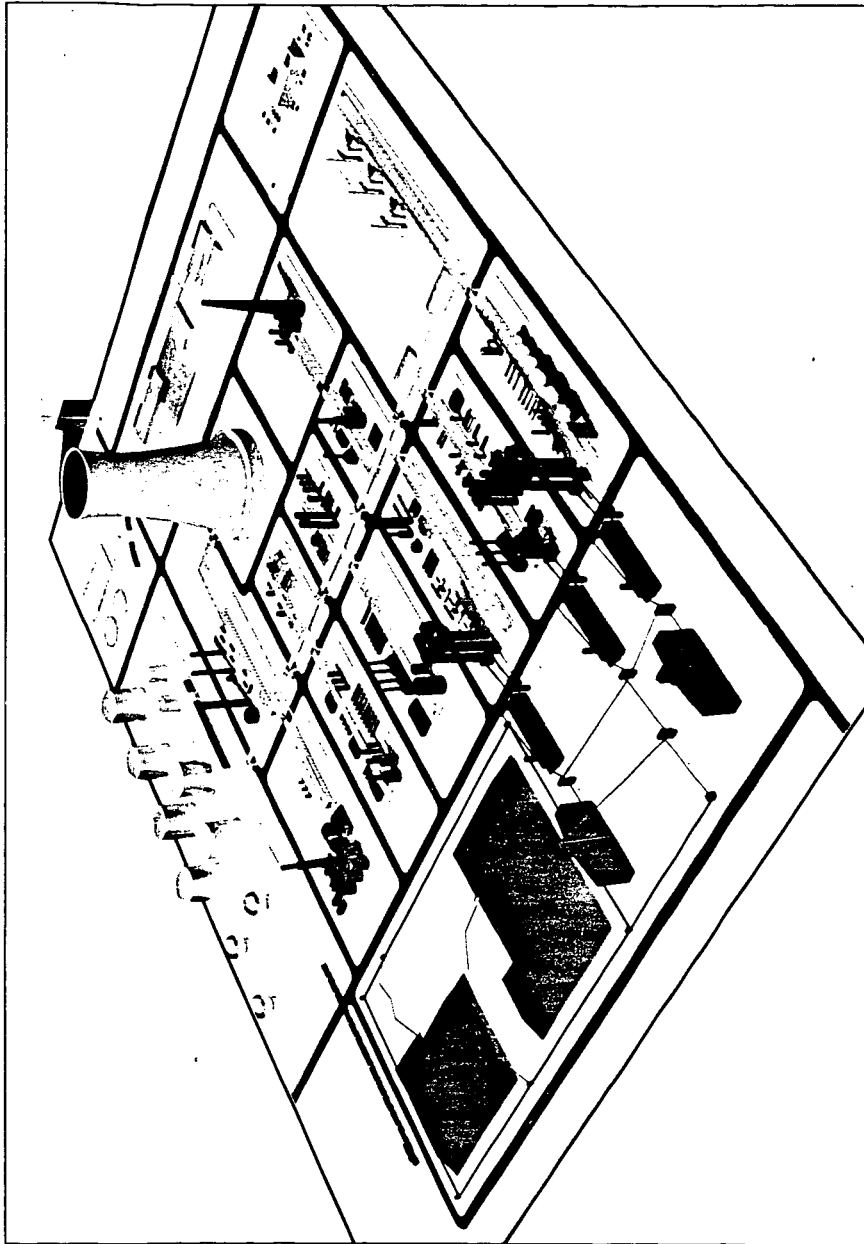


Figure 2 - Model of Conceptual Oil/Gas Plant Design

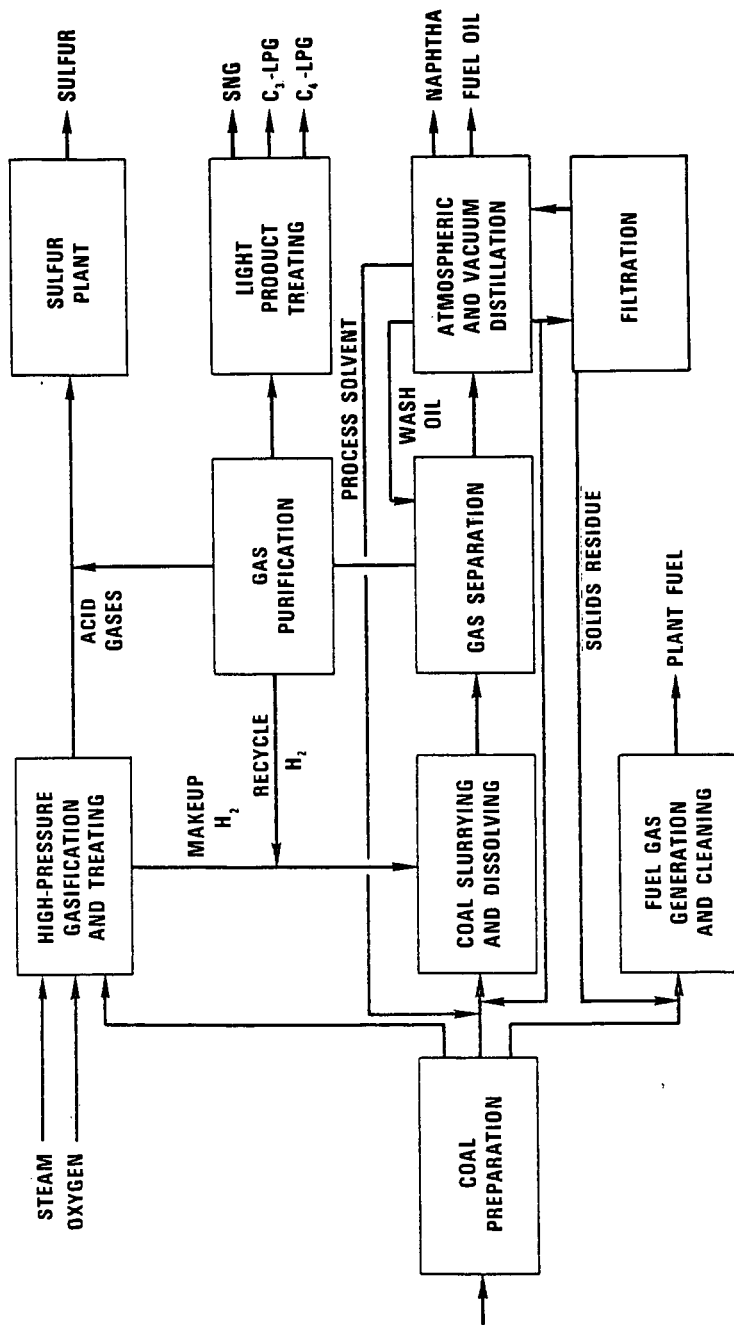


Figure 3 - Oil/Gas Plant
Simplified Block Flow Diagram

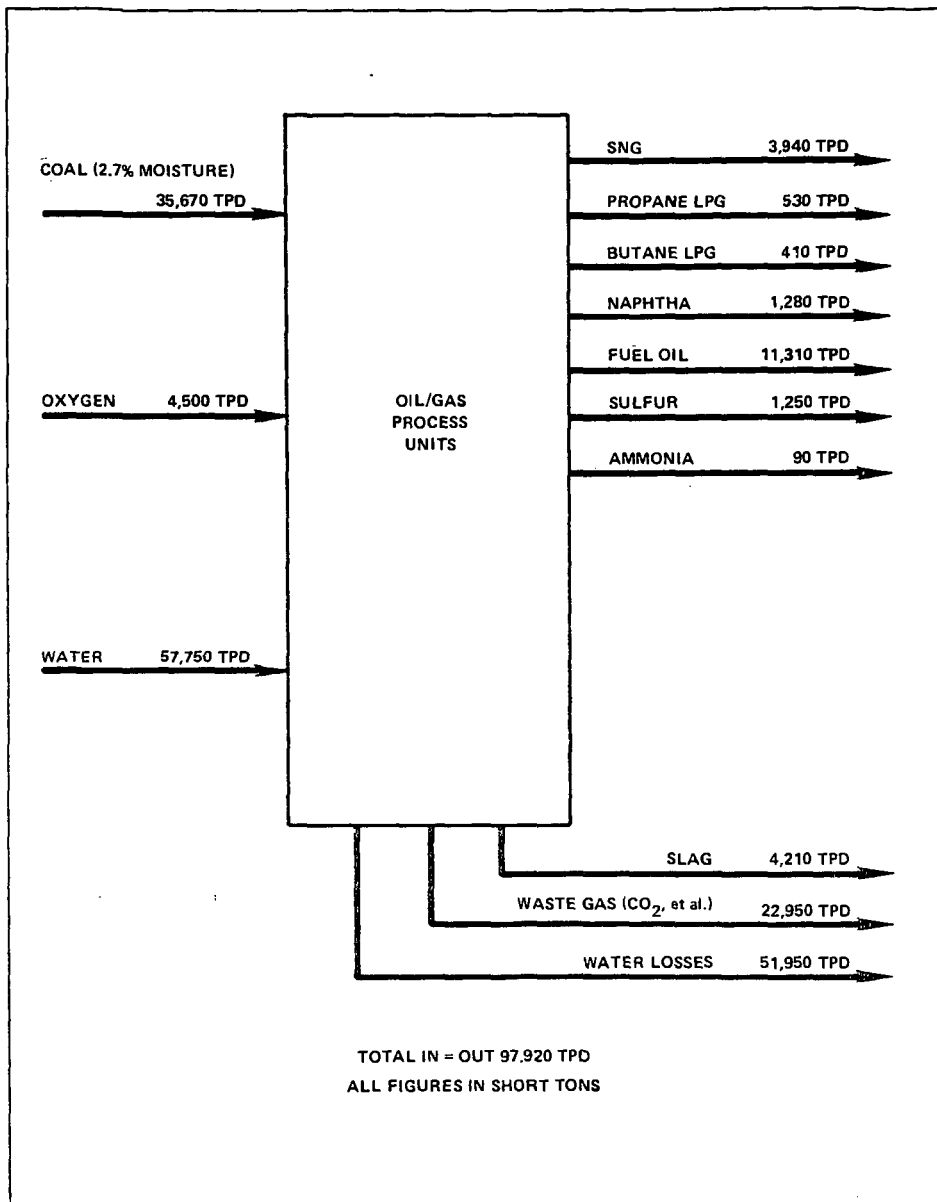
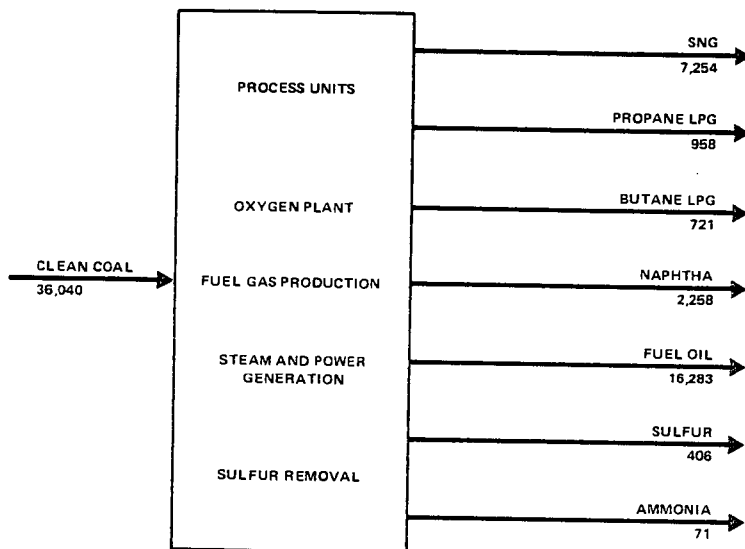


Figure 4 - Overall Material Balance
Oil/Gas Plant



ALL FIGURES ARE MM BTU/HR, HHV

$$\text{THERMAL EFFICIENCY} = \frac{7,254 + 958 + 721 + 2,258 + 16,283 + 406 + 71}{36,040} = 77.6\%$$

Figure 5 - Thermal Efficiency, Oil/Gas Plant

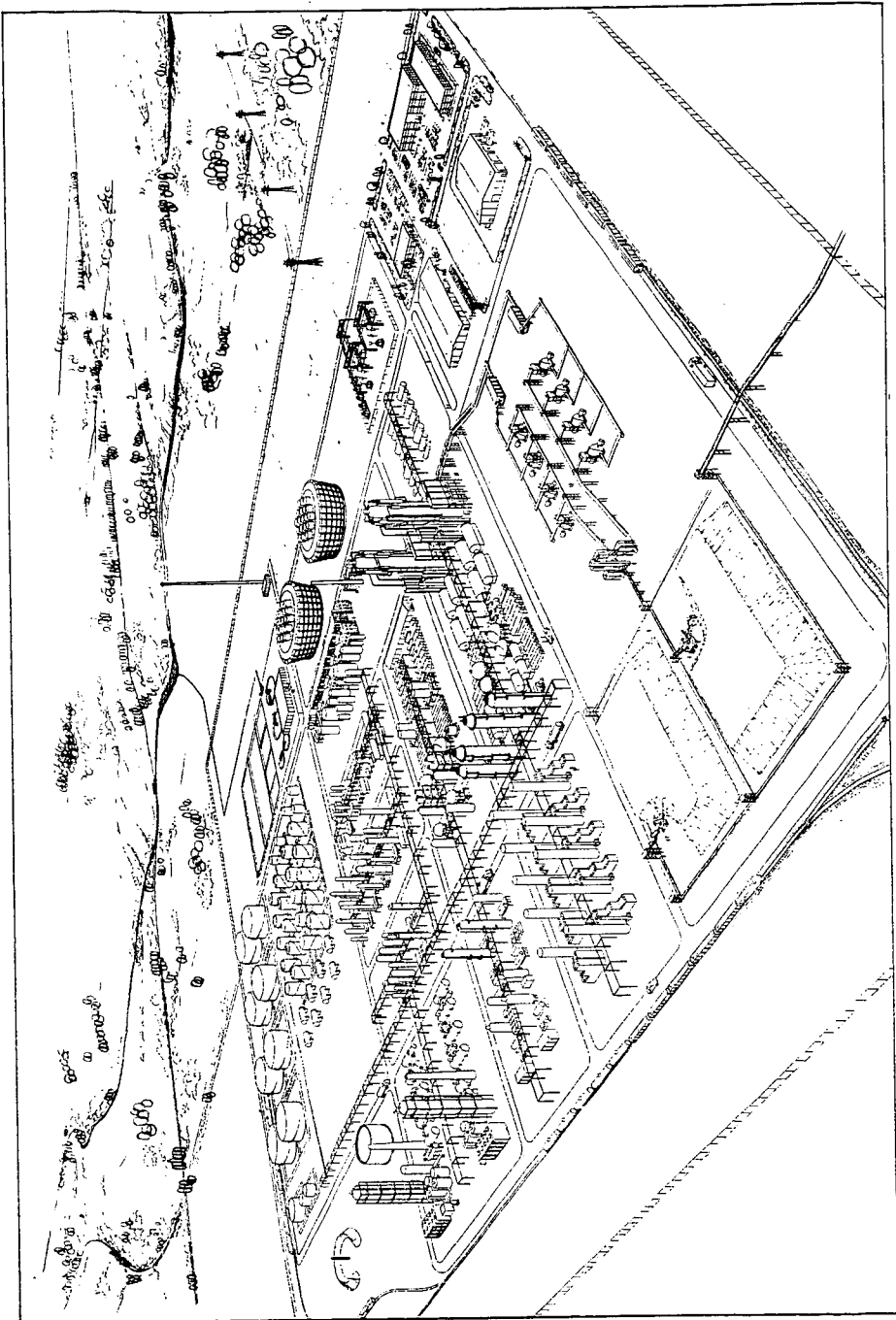


Figure 6 - Artist's Conceptual Drawing
Fischer-Tropsch Plant

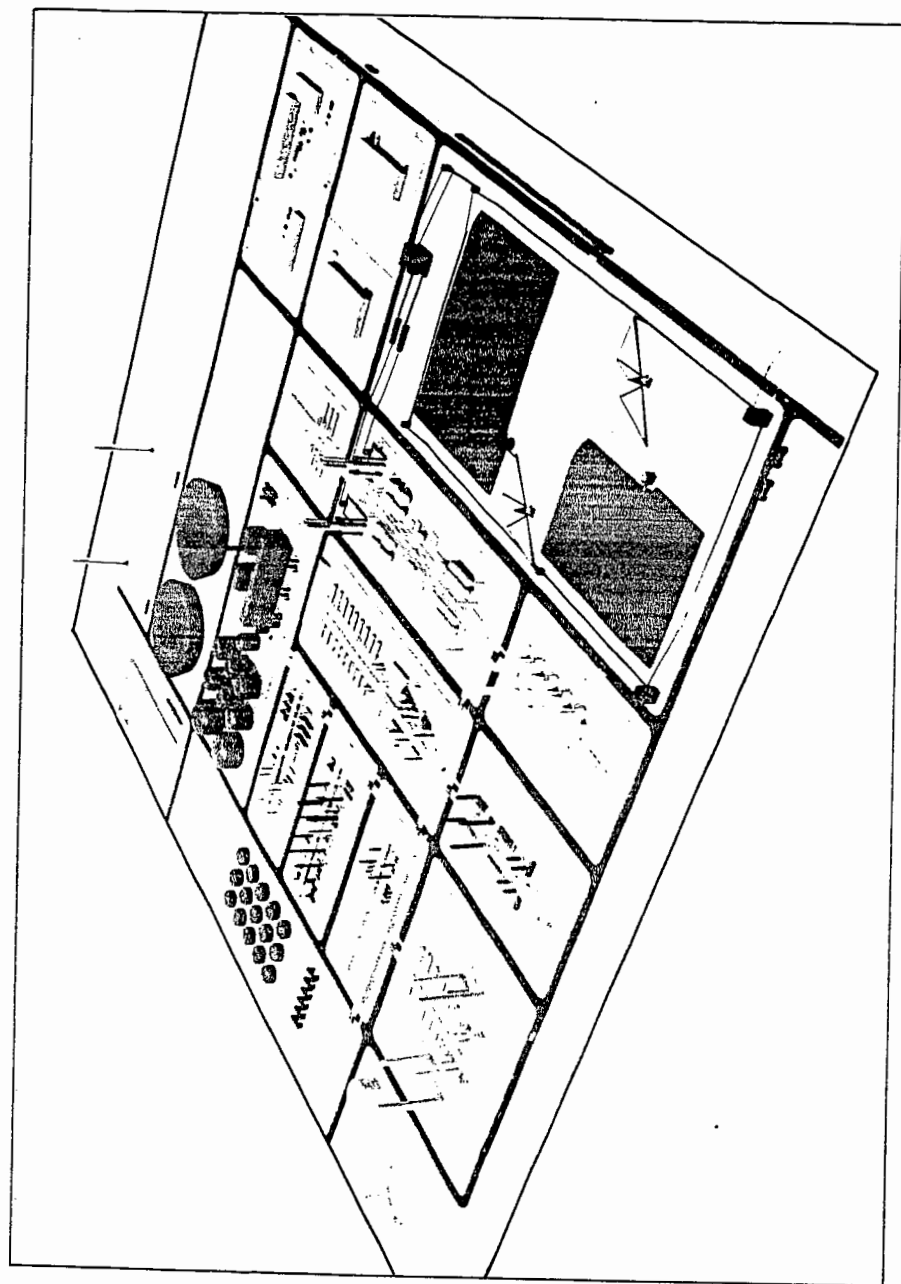


Figure 7 - Model of Conceptual Fischer-Tropsch
Plant Design

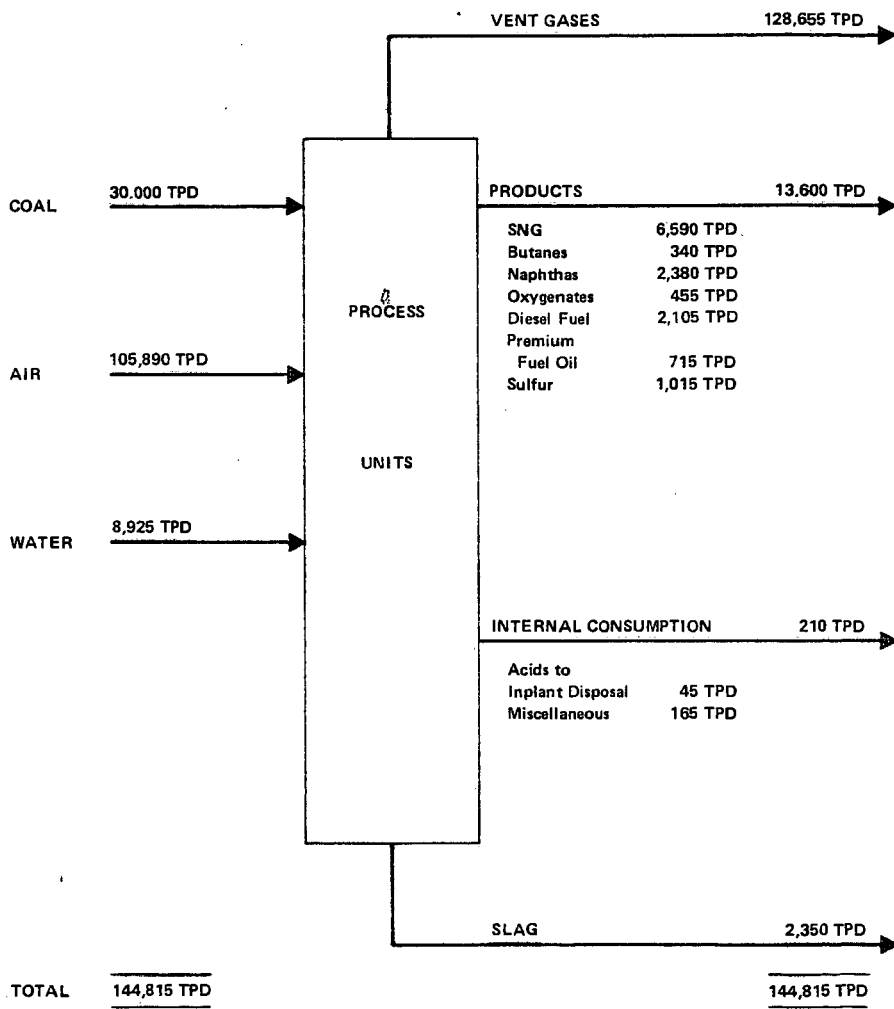
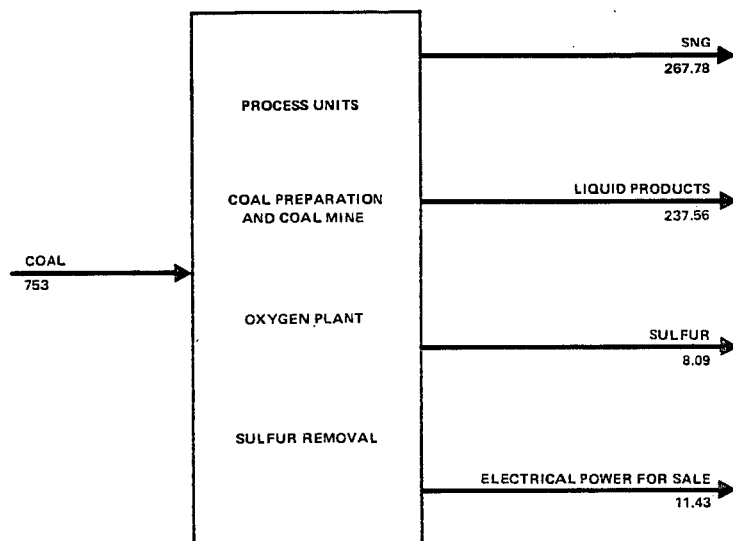


Figure 8 - Overall Material Balance
Fischer-Tropsch Plant



ALL FIGURES ARE MMM BTU/D, HHV

$$\text{THERMAL EFFICIENCY} = \frac{267.78 + 237.56 + 8.09 + 11.43}{753} = 69.7\%$$

Figure 9 - Thermal Efficiency
Fischer-Tropsch Plant

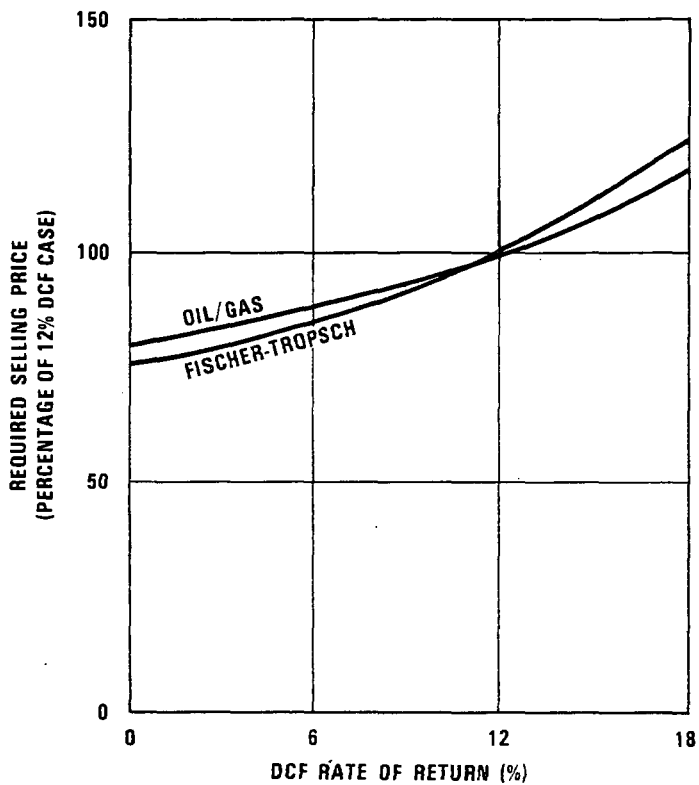


Figure 10 - Sensitivity of Required Product
Selling Price to DCF, 65% Debt Case

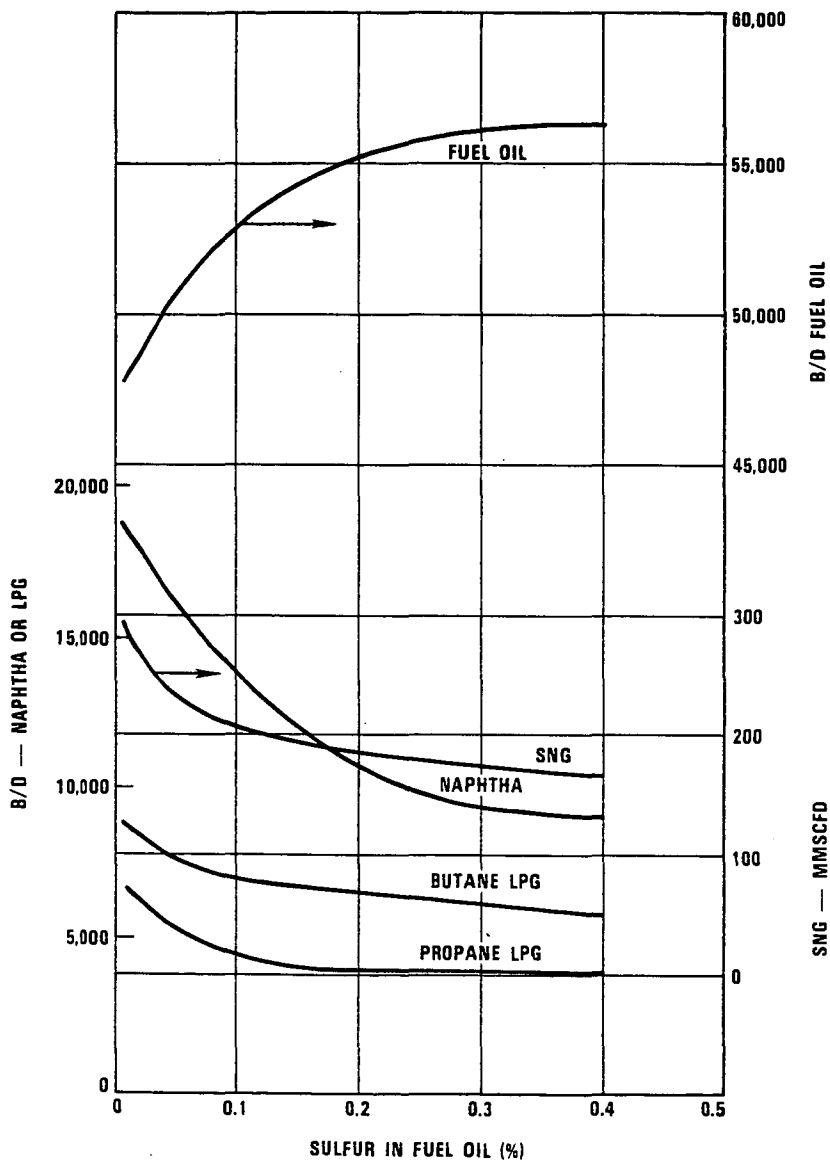


Figure 11 - Projected Product Distribution
Product Yield vs. Fuel Oil Sulfur Content
Oil/Gas Plant

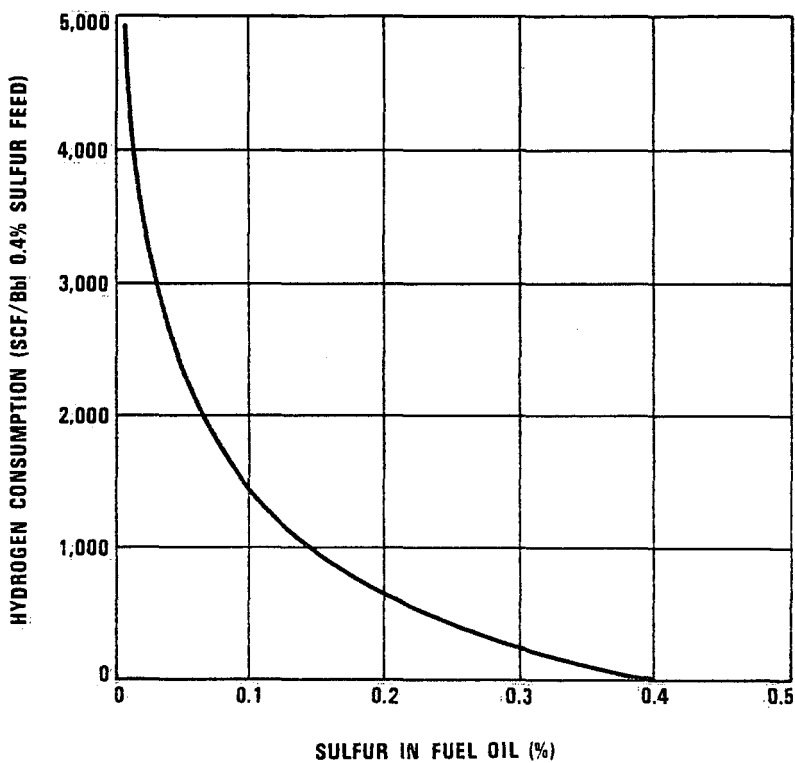


Figure 12 - Projected Hydrogen Consumption
Hydrogen Consumed vs. Fuel Oil Sulfur Content
Oil/Gas Plant

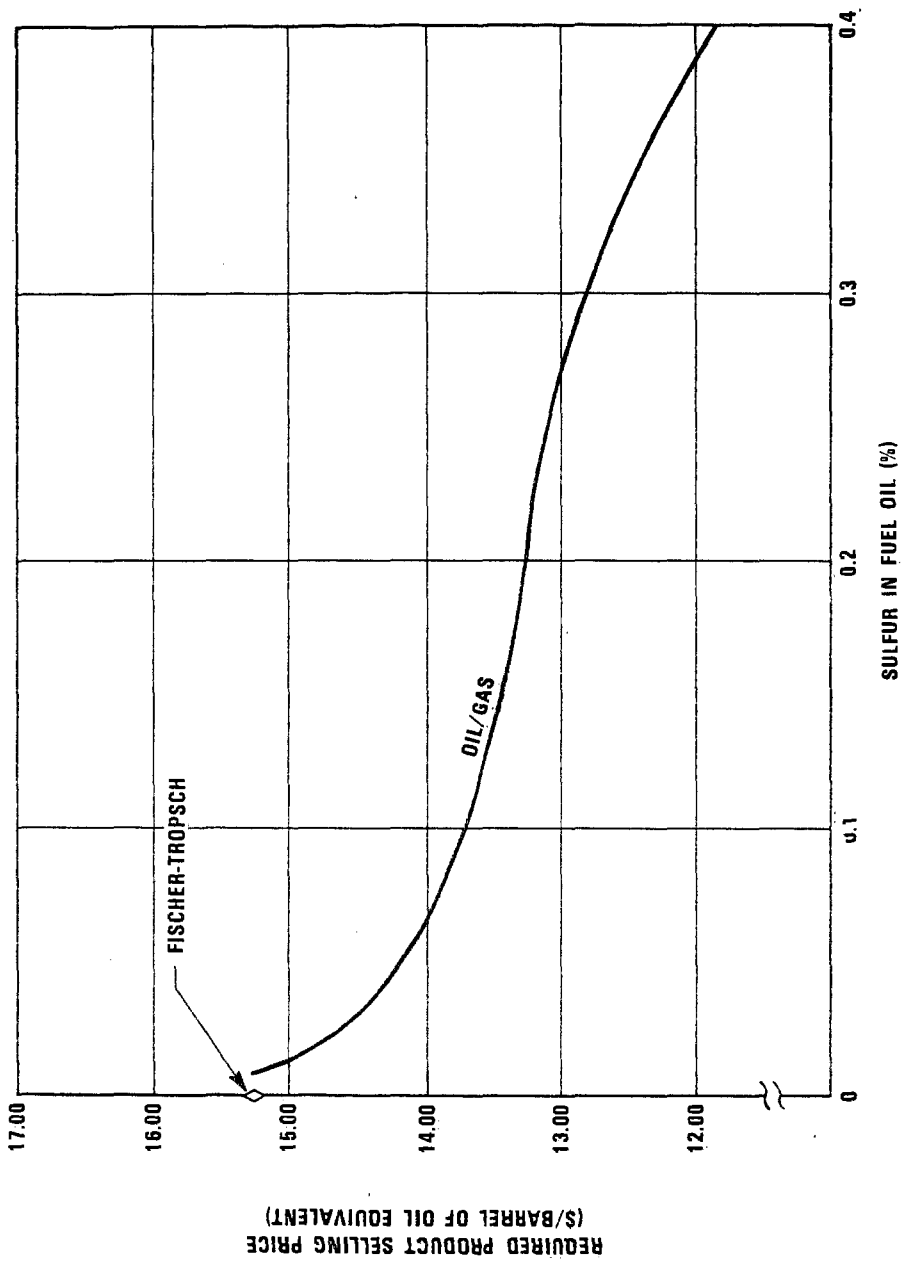


Figure 13 - Required Average Product Selling Price as a Function of the Sulfur Content of Oil/Gas Fuel Oil

Table I. Comparison of Projected Product Characteristics

Product	Projected Characteristics	
	Fischer-Tropsch	Oil/Gas
SNG	Pipeline Quality	Pipeline Quality
C ₃ LPG	--	Propane 210 psia Vapor Pressure
C ₄ LPG	Mixed Butane - Butylene 37 psia Vapor Pressure	Mixed Propane-Butane, 70 psia Vapor Pressure
Full Range Naphtha	--	50° API Gravity C ₅ to 380°F ASTM EP High Naphthene
Light Naphtha	Nil Sulfur 185°F ASTM EP 85.5°API Gravity	--
Heavy Naphtha	Nil Sulfur 300°F ASTM EP 71.3 API Gravity	--
Diesel Fuel	57°API Gravity 60 plus Cetane Number Nil Sulfur, Nil Nitrogen	--
Fuel Oil	41°API Gravity Nil Sulfur Higher Heating Value: 19,900 Btu/lb	-8.2°API Gravity 0.4 wt % Sulfur Higher Heating Value: 17,200 Btu/lb

Table II. Comparison of Fixed Capital Investments (FCI) for Fischer-Tropsch and Oil/Gas Complexes

Barrels Fuel Oil Equivalent/Day (BPOE/D): F-T = 86,000
O/G = 110,000

Description	Fischer-Tropsch \$ Millions	Oil/Gas \$ Millions
Mine and Coal Preparation		
Mine	175.6	211.6
Coal Preparation	22.0	30.0
Coal Storage	11.2	13.0
Crushing and Drying	13.0	15.1
Subtotal	221.8	269.7
Conversion		
Fischer-Tropsch Synthesis	204.6	--
Oil Recovery and Fractionation	30.5	--
Chemical Recovery	15.9	--
Slurry and Dissolving	--	216.8
Filtration	--	42.0
Distillation	--	31.6
Dissolver Acid Gas Removal	--	20.3
Subtotal	251.0	310.7
Process Gas Production		
Gasification	37.3	45.4
Heat Rec. and Part. Removal	151.2	--
Acid Gas Removal	100.3	47.7
Shift	18.9	59.5
Power Generation	119.6	86.8
Subtotal	427.3	239.4
SNG Separation and Treatment		
Methanation	60.6	0.6
SNG and LPG Treating	--	48.3
Subtotal	60.6	48.9
Product Finishing		
Sulfur Plant	22.1	15.4
Naphtha Hydrogenation	--	9.2
Subtotal	22.1	24.6
Utilities		
Oxygen Plant	305.3	90.2
Instrument and Plant Air	3.6	2.4
Potable and Sanitary Water	0.4	--
Raw Water System	23.8	--
Fuel Gas Gasify	--	71.2
Fuel Gas Acid Gas Removal	--	17.9
Raw Water Treating	--	16.5
Subtotal	335.1	198.2
Environmental and General Facilities		
General Facilities	19.5	37.2
Water Reclaiming	40.4	--
Effluent Water Treating	3.0	5.5
Product Storage	31.2	32.2
Sour Water Stripping	--	5.9
Subtotal	84.1	80.8
Total Constructed Cost	1,420.0	1,172.3
Home Office Costs	140.2	117.2
Sales Tax	28.1	23.5
Total Fixed Capital Investment (FCI)	1,570.3	1,315.0
FCI/(BPOE/D)	18,250	11,950

Table III. Comparison of Relative Fixed Capital Investments of Fischer-Tropsch and Oil/Gas by Unit

Description	Ratio of Fischer-Tropsch to Oil/Gas	
	Fixed Capital Investment	FCI/BOE
Mining & Coal Preparation	0.82	1.05
Conversion	0.81	1.03
Process Gas Production	1.78	2.28
SNG Separation & Treatment	1.24	1.58
Product Finishing	0.90	1.15
Utilities	1.69	2.16
Environmental & General Facilities	<u>1.04</u>	<u>1.33</u>
Total	1.19	1.53

Table IV. Comparison of Projected Total Capital Requirements for Fischer-Tropsch and Oil/Gas Complexes

Item	Fischer-Tropsch \$ MM	Oil/Gas \$ MM	Ratio F-T - O/G
Fixed Capital Investment	1550	1300	1.19
Initial Catalyst & Chemicals	11	9	1.22
Start-Up Costs	110	86	1.28
Construction Financing ^a	212	188	1.13
Working Capital	113	107	1.06
Land, Rights of Way	<u>1</u>	<u>1</u>	<u>1.00</u>
TOTAL	1997	1691	1.18
Say	<u>2000</u>	<u>1700</u>	
a) Example: For 65/35 debt/equity, 9% interest, 0.75% commitment fee case.			

Table V. Comparison of Projected Annual Operating Costs for Fischer-Tropsch and Oil/Gas Complexes

Cost Center	Annual Operating Costs - \$MM	
	Fischer-Tropsch	Oil/Gas
Coal Mine	84.5	104.2
Coal Preparation	2.3	3.2
Process Plant	101.5	84.4
Power Plant	7.7	--
Offsites	<u>7.8</u>	<u>14.4</u>
TOTAL	203.8	206.2
Say	<u>205</u>	<u>205</u>

Table VI. Comparison of Projected Average Required Product Selling Price at 12% DCF

Project Financial Structure	Required Average Product Selling Price in Dollars per Million BTU		
	Fischer-Tropsch	Oil/Gas	Ratio F-T - O/G
100% Equity	3.30	2.50	1.32
65/35 Debt/Equity	2.55	1.95	1.28
Breakeven	1.50	1.20	1.20

Table VII. Sensitivities of Average Required Product Selling Price to Key Economic Parameters

Economic Parameter	Sensitivity of Average RPSD, %			
	Fischer-Tropsch		Oil/Gas	
	100% Equity	65% Debt	100% Equity	65% Debt
Fixed Capital Investment	87	81	82	78
Operating Costs	15	19	21	27
Run of Mine Coal Costs	21	25	30	34

Table VIII. Possible Product Sales Values for Fischer-Tropsch Complex

Product	Daily Production	Possible Unit Sales Value in Dollars	Annual Gross Revenue in \$ Million
<u>SNG</u>	260.0 MMscfd	4.25/Mcf	362.8
<u>Liquids</u>			
C ₄ s	3,535 BPD	12.00/bbl	14.0
<u>Naphthas</u>			
Light	10,620 BPD	15.50/bbl	54.3
Heavy	9,555 BPD	17.00/bbl	53.6
Alcohols	3,910 BPD	25.00/bbl	32.3
Diesel Fuel	16,960 BPD	14.50/bbl	79.9
Premium Fuel Oil	4,960 BPD	15.00/bbl	<u>24.5</u>
			241.6
<u>Power</u>	3,352 MW/hr	0.03/kW-hr	<u>33.2</u>
Total Energy			651.6
Sulfur	1,015 Ton	60/ton	<u>20.1</u>
Total			671.7
4th Qtr. 1975			
Escalation			60.5
(9% from 4th Qtr. 1975 to 4th Qtr. 1976)			
Total			
4th Qtr. 1976			<u>732.2</u>

Table IX. Possible Product Sales Values
for Oil/Gas Complex

Product	Daily Production	Possible Unit Sales Value in Dollars	Annual Gross Revenue in \$ Million
SNG	170 MMscfd	4.25/Mcf	238.425
Propane	6,030 BPD	11.00/bbl	21.890
Butane	4,100 BPD	12.00/bbl	16.235
Naphthas	9,400 BPD	15.50/bbl	48.080
Fuel Oil	56,400 BPD	9.75/bbl	<u>181.470</u>
Total Energy			506.100
Sulfur	118 LT/D	60/ton	2.335
Ammonia	90 ST/D	120/ton	<u>5.565</u>
Total 4th Qtr. 1975			514.000
Escalation (9% from 4th Qtr. 1975 to 4th Qtr. 1976)			46.000
Total 4th Qtr. 1976			<u><u>560.000</u></u>

Table X. DCF's for Possible Product Revenues

Project Financial Structure	DCF	
	Fischer-Tropsch	Oil/Gas
100% Equity	17	13
65/35 Debt/Equity	27	20

COSTS OF SOLIDS-LIQUIDS SEPARATION METHODS IN COAL LIQUEFACTION

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One of the major process steps in the conversion of coal to oil is the separation of the residual char or ash from the product liquid. Internal studies by Dravo have shown the cost of such separations to vary from 5 to 20% of the total product oil cost. Estimates of cost of coal derived oil range from \$15 to \$25 per bbl⁽⁷⁾ and upward providing a high incentive to recover a maximum of the oil associated with solids. Furthermore, the solids content of the product oil must not exceed .1% by weight if the oil is to be used as fuel so that the users can burn it without installing precipitators on their flue gas stacks. Also, if the oils are to be hydrotreated, solids levels less than 0.1 percent are required since fines from catalyst attrition would combine with residual solids and cause the final product to exceed this residual solids specification⁽¹⁾. Because of this separation specification, the severe operating conditions, and the propensity for plugging, coking, etc., the number of reliable process schemes is rather limited.

Dravo selected a typical liquefaction process, generally similar to Synthoil, and examined a number of solids removal systems in an attempt to find a reliable, cost effective scheme.

In the selected liquefaction process, part of the liquid product is recycled and used to slurry the coal feed to the liquefaction reactor. This feed slurry can utilize a recycle stream which has been treated to reduce its solids content from 12 to 6 weight percent. This is accomplished in a bank of hydroclones. The hydroclone feed, at 400 psig and 560°F, is split into two streams - the overheads, which is recycled to the feed preparation system, and the bottoms, which exits at 15 weight percent solids and 240 psig. This stream must now be treated further.

Several methods of secondary separation were investigated. Tests on hydroclones and centrifuges have not demonstrated the required solids removal efficiencies. Precoat filtration, on the other hand, has been successfully tested⁽²⁾. The high rates obtained when filtering the oils produced in this selected process, when compared to SRC and COED filtration rate data, increases the attractiveness of filtration. This high rate is due in part to the comparatively large amount of hydrogen consumed in the liquefaction reactor, resulting in a lower viscosity of the product oil. A preliminary screening indicated that filtration at these higher rates is comparable to other separation methods on a capital cost basis⁽¹⁾. For these reasons, it was decided to include pressure precoat filtration in the economic tradeoff analysis.

Secondary separation can also be carried out by feeding the hydroclone bottoms to the base of the product fractionator. Proper baffling should produce a sufficiently tortuous path to allow most of the solids to remain in the bottoms. This is not unlike the oil absorption tests run on COED oils, in which the majority of the solids carryover was removed in the first contact stage (bottoms), while the remaining, lighter fractions were recovered relatively solids-free⁽³⁾.

As mentioned earlier, economics dictate that essentially all the oil be recovered from the sludge produced in the secondary separation step. The oil contained in the sludge amounts to about 20% of the production rate. Some oil diffuses into the extraction solid (char) pores and remains there through capillary action. Solvent extraction or heat treatment is required to recover this oil⁽⁴⁾. Solvent extraction is currently in the development stage and requires an extra separation step⁽⁵⁾. Low pressure fluid bed dryers similar to those used in Project Gasoline were chosen as a viable method of effecting complete separation. Oil loss by coking is estimated at

4 percent of the oil fed to the dryer. The recovered char is pneumatically conveyed to a gasifier and used as hydrogen production feedstock.

Three alternate separation systems were decided on as a result of this initial screening. Case 1 (Figure 1) employs a hydroclone-rotary pressure precoat filter-fluid bed dryer solids separation sequence. The filter feed is at 200 psig and 500°F. In Case 2 (Figure 2), the filtering step is eliminated, increasing the capacity of the fluid bed dryer equipment. In Case 3 (Figure 3), the hydroclone underflow is fed directly to the base of the fractionator, which is baffled for the removal of the solids with the bottoms. The final liquid-solids separation, as previously stated, is carried out in the fluid bed dryer section. To keep the cost comparison on a consistent base, fractionation charges were included for all three cases.

Solids separation costs for this 50,000 Bbl/day facility were calculated by the Discounted Cash Flow method, using the following basis: 20-year project life, 16-year sum-of-the-years-digits depreciation on total Plant Investment, 100 percent equity capital, 12 percent DCF return rate, and 48 percent federal income tax rate⁽⁶⁾.

In addition, the following unit costs were employed in determining the annual operating costs:

Low Pressure Steam	\$2.30/MM Btu
Medium Pressure Steam	\$2.50/MM Btu
Process Water	\$.40/M Gal
Cooling Water	\$.03/M Gal
Electric Power	\$.025/KW/HR
Fuel Gas	\$3.50/MM Btu
Operating Labor	\$15,000/man/year
Maintenance Charges	3% of total installed cost for oil absorption and fractionation areas 6% of total installed cost for hydroclone, fluid bed dryer, and drum filter areas

The following results were obtained:

	<u>CASE 1</u>	<u>CASE 2</u>	<u>CASE 3</u>
Installed Cost, \$M			
Hydroclone Area	16021	16021	16021
Drum Filter Area	25039	---	---
Fluid Bed Dryer Area	13002	39332	15665
Oil Absorption Area	2321	5635	2702
Fractionation Area	12908	12908	13153
Total Installed Cost, \$M	69291	73896	47541
Annual Operating Cost \$M/yr	25100	43566	21718
Total Separation Charges, \$/BBL	2.66	3.83	2.14

The results show Case 3 to be the least costly separations method. This seems reasonable since two operations, secondary solids separation and fractionation, are combined. Case 1 is somewhat more expensive, and would be more competitive if a higher filtration rate could be obtained. Case 2, however, is much more costly than either of the other alternates. This is mainly due to the higher capital costs required in the Fluid Bed Dryer area and the accompanying large increase in fuel gas usage.

In summary, the most economical of the liquid-solids separations methods analyzed appears to be Case 3, the combined secondary separation-fractionation alternate. Pilot tests would be recommended prior to including this system as part of a commercial facility.

OPERATING COST SUMMARY CASE I

	<u>\$M/YR</u>
<u>CATALYSTS AND CHEMICALS</u> - Filter Aid	676
<u>UTILITIES</u> - Steam	2,154
Process Water	211
Cooling Water	317
Electric Power	1,493
Fuel Gas	11,304
<u>LABOR</u> Operating	600
Maintenance	2,220
Supervision	564
<u>ADMINISTRATION AND GENERAL OVERHEAD</u>	2,030
<u>SUPPLIES</u> - Operating	180
Maintenance	1,480
<u>LOCAL TAXES AND INSURANCE</u>	1,871
<u>TOTAL GROSS OPERATING COSTS</u>	25,100
<u>TOTAL NET OPERATING COSTS</u>	25,100

OPERATING COST SUMMARY CASE II

	<u>\$M/YR</u>
<u>UTILITIES</u> - Steam	1,649
Process Water	359
Cooling Water	540
Electric Power	2,818
Fuel Gas	29,389
<u>LABOR</u> Operating	360
Maintenance	2,326
Supervisory	537
<u>ADMINISTRATION AND GENERAL OVERHEAD</u>	1,934
<u>SUPPLIES</u> - Operating	108
Maintenance	1,551
<u>LOCAL TAXES AND INSURANCE</u>	1,995
<u>TOTAL GROSS OPERATING COSTS</u>	43,566
<u>TOTAL NET OPERATING COSTS</u>	43,566

OPERATING COST SUMMARY CASE III

	<u>\$M/YR</u>
<u>UTILITIES</u> - Steam	1,649
Process Water	199
Cooling Water	299
Electric Power	955
Fuel Gas	12,844
<u>LABOR</u> Operating	360
Maintenance	1,426
Supervision	357
<u>ADMINISTRATION AND GENERAL OVERHEAD</u>	1,286
<u>SUPPLIES</u> Operating	108
Maintenance	951
<u>LOCAL TAXES AND INSURANCE</u>	1,284
<u>TOTAL GROSS OPERATING COSTS</u>	21,718
<u>TOTAL NET OPERATING COSTS</u>	21,718

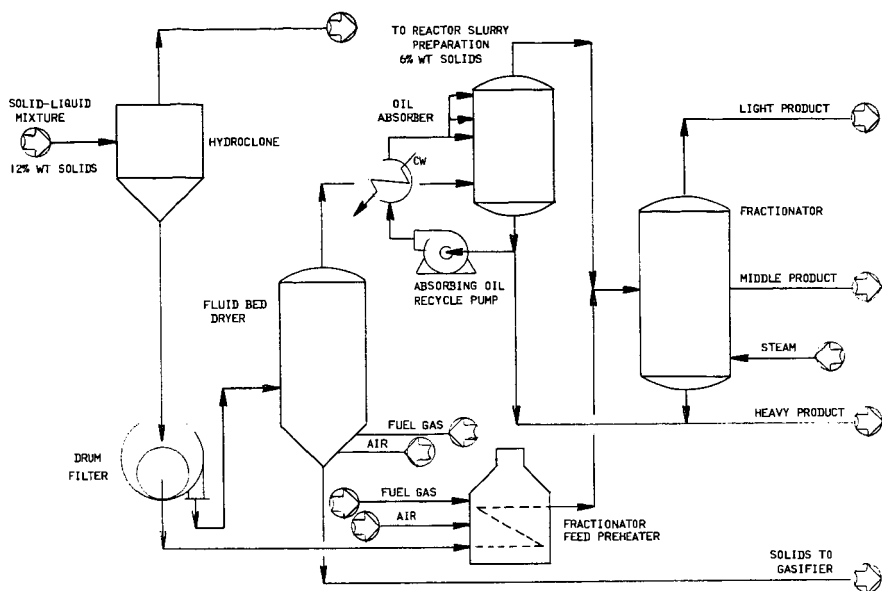


FIG.1 PRESSURE FILTRATION CASE I

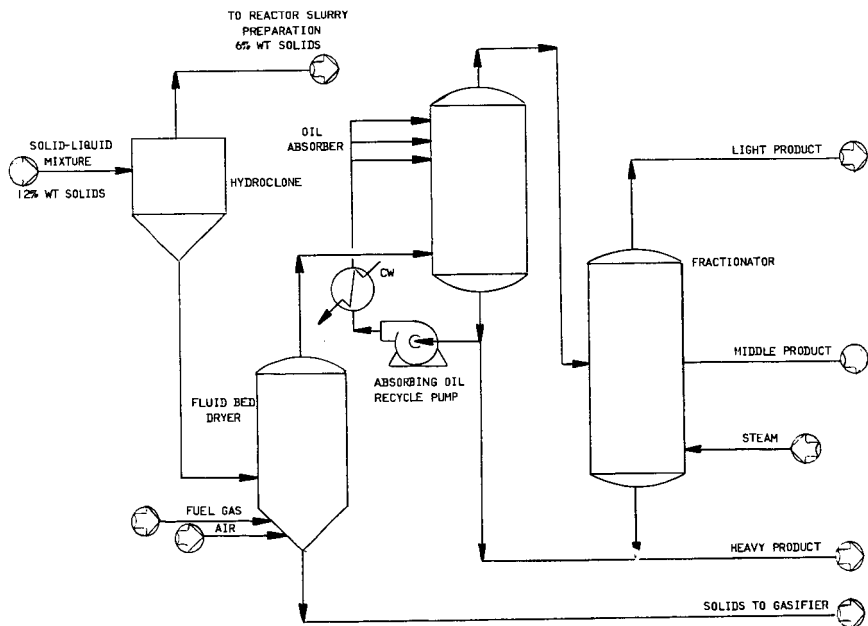


FIG.2 FLUID BED DRYER CASE II

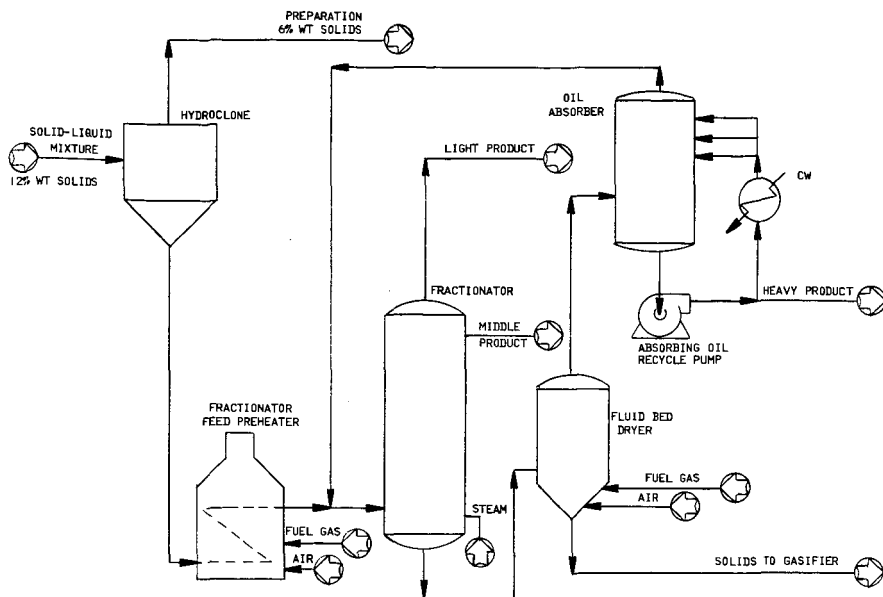


FIG.3 FRACTIONATION CASE III

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REGIONAL AND FEEDSTOCK EFFECTS ON ECONOMICS OF INTEGRATED COAL GASIFICATION/POWER PLANT SYSTEMS

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INTRODUCTION

Coal's major role in alleviating our energy shortage depends on our ability to derive clean fuels from it. Low and medium Btu gas from coal can be important industrial and utility fuels. We are presenting the results of an investigation into how competitive these coal derived fuels are for power generation. Coal gasifiers integrated with either combined cycle or conventional steam cycle power plants are compared with conventional coal fired power plants with and without flue gas desulfurization (FGD).

The geographical areas selected for study purposes are two National Electric Reliability Council (NERC) Regions--the Chicago area (MAIN Region) and the New England Area (NPCC Region). In the MAIN region, the high sulfur coals studied were Illinois No. 6, an eastern coal, and Rosebud, a western coal. The low sulfur coals studied were Stockton, West Virginia, eastern coal and Wyodak western coal. The same coals were used for the NPCC region except that Middle Kittanning coal was the representative high sulfur eastern coal. The characteristics of coal selected are summarized in Table 1.(1)

BASIS OF POWER PLANT DESIGN

800 Mw is the base load unit size in this study. Capacity factor is 70%. Coal storage and handling facilities provide capacity for 60 days onsite storage.

For a combined cycle base load unit, the study case plant contained four 200 Mw modules, each consisting of a gas turbine, heat recovery boiler, steam turbine, and generator.

The fixed capital costs for all power plant configurations and fixed operating costs for the two conventional power plant technologies are summarized in Table 2. The fixed operating costs for the integrated cases are discussed in a separate section.

GASIFIER SELECTION

Selection Criteria

Although gasifiers differ in many ways, they are generally classified according to coal flow within the reactor. In a fixed-bed gasifier, the steam required for grate cooling and for preventing clinker formation is greater than the amount of steam required for the gasification reaction, thereby lowering the overall thermal efficiency in gasification. In addition, due to the large coal particle sizes and the moderate temperature involved, the fixed-bed gasification rates are low, and solid residence times of one to two hours are required. These gasifiers, however, have excellent turndown capabilities.

In a fluidized bed, the upward flow of gas is at a velocity slightly above that required to merely support the coal. The relatively short coal residence time (20 to 40 min.) results in a lower operating efficiency than for the fixed bed. Increasing the thermal efficiency requires increasing the coal residence time by using multistage beds to obtain the countercurrent conditions.

TABLE 1. Characteristics of Coals Selected

	High Sulfur Coal			Low Sulfur Coal	
	<u>Illinois No. 6</u>	<u>Rosebud</u>	<u>Mid-Kittanning</u>	<u>Stockton</u>	<u>Wyodak</u>
<u>Proximate Analysis: %</u>					
Moisture	9.7	9.8	3.3	3.0	29.5
Volatile Matter	36.6	35.2	30.1	34.9	30.1
Fixed Carbon	42.2	46.7	57.5	54.3	33.9
Ash	<u>11.5</u>	<u>8.3</u>	<u>9.1</u>	<u>7.8</u>	<u>6.5</u>
Total	100.0	100.0	100.0	100.0	100.0
<u>Ultimate Analysis: %</u>					
Hydrogen	5.3	5.2	5.2	5.2	7.3
Carbon	63.4	60.8	75.3	75.4	45.7
Nitrogen	1.4	0.9	1.3	1.4	1.1
Oxygen	13.9	22.8	6.9	9.6	39.0
Sulfur	4.5	2.0	2.2	0.6	0.4
Ash	<u>11.5</u>	<u>8.3</u>	<u>9.1</u>	<u>7.8</u>	<u>6.5</u>
Total	100.0	100.0	100.0	100.0	100.0
<u>HHV, Btu/lb</u>	11,605	10,379	13,282	13,084	8,167
<u>Ash Fusibility, °F</u>					
Initial	2,330	2,010	2,020	2,910+	2,163
Softening	2,430	2,060	2,080	2,910+	2,223
Fluid	2,590	2,110	2,210	2,910+	2,250

In an entrained bed, the raw coal fed into the unit is transported by the velocity of the gas. The extent of coal conversion to gas is limited by the short solid residence time of less than ten seconds. In order to achieve essentially complete conversion and to maintain high thermal efficiency, a multistage countercurrent unit is desired.

For application to power plants of both conventional and combined cycle type, a gasification process with a high throughput and a high degree of reliability is desirable. Gasifier turndown capability is of less importance for base load units. In a combined cycle, high pressure gasifiers are desirable, whereas low pressure gasifiers are satisfactory for conventional cycles.

A review of gasifier specifications indicates that an entrained-bed gasifier meets the criteria, i.e., pressurized, single stage for combined cycle applications and low pressure, two-stage for conventional cycle power plant applications.

TABLE 2. SUMMARY OF POWER PLANT FIXED CAPITAL AND OPERATING COSTS

BASIS: JANUARY 1977

	MAIN				NPCC			
	HIGH SULFUR COAL		LOW SULFUR COAL		HIGH SULFUR COAL		LOW SULFUR COAL	
	TILL NO. 6	ROSEBUD	STOCKTON	WYODAK	MID. KITITANING	ROSEBUD	STOCKTON	WYODAK

FIXED CAPITAL COST: MM\$

LOW SULFUR W/O FGD	-	-	420.4	432.8	-	-	424.6	437.1
HIGH SULFUR W/FGD	547.8	547.0	-	-	550.8	552.4	-	-
MED. BTU W/COMB. CYC.	227.4	227.4	227.4	227.4	229.7	229.7	229.7	229.7
LOW BTU W/COMB. CYC.	227.4	227.4	227.4	227.4	229.7	229.7	229.7	229.7
MED BTU W/CONV. CYC.	277.6	277.6	277.6	277.6	280.4	280.4	280.4	280.4
LOW BTU W/CONV. CYC.	280.7	280.7	280.7	280.7	283.5	283.5	283.5	283.5

FIXED OPERATING COST: MM\$/YR

LOW SULFUR W/O FGD	-	-	6.2	6.3	-	-	6.3	6.4
HIGH SULFUR W/FGD	20.9	20.9	-	-	21.2	21.1	-	-

Fuel Characteristics

The different gasification processes produce variations in raw or clean gas composition. However, for a given gasification process, experimental data using various coal feeds, ranging from bituminous to lignite, indicate that the characteristics of clean or raw gas composition are almost independent of types of coal employed. For example, the raw gas composition from low pressure two-stage, oxygen-blown, entrained-bed gasifiers does not vary greatly when fed with bituminous, subbituminous or lignite⁽²⁾. For the present study, therefore, it is assumed that the product gas composition from a selected gasifier is independent of the type of coal feed. Typical fuel characteristics for low and medium Btu gas obtained from an entrained gasifier are presented in Table 3.

Process Description

Simplified block flow diagrams of the integrated gasification/conventional boiler and combined cycle plants are shown in Figures 1 and 2, respectively. Since the entrained-bed gasifier was selected for the applications of both power plant configurations, the gasification process description presented is valid for both power plant applications.

In the entrained-bed gasifier, prepared, pulverized coal is fed to the gasifier along with steam and oxygen/air. Low pressure steam for the gasifier reaction is produced in the gasifier cooling jacket. Raw gas at 2700°F is usually water quenched and then passed through a waste heat boiler. The gas is cooled in a venturi scrubbing system and sent to a suitable desulfurization system. The clean product gas is then sent to the fuel ports of the steam generator (boiler). A balanced-draft, tangentially-fired, controlled circulation steam generator is used to burn the clean, low or medium Btu gas.

For the combined cycle facility, compressed air and cleaned fuel gas are fired in the combustion chamber of the gas turbine. The hot combustion gases are then expanded through the turbine to generate electrical power. The exhaust from the gas turbine is used further to generate high pressure steam in an unfired boiler before being sent to the stack. The high pressure steam drives the steam turbine to generate additional electric power.

BASIS OF INTEGRATED GASIFIER/POWER PLANT SYSTEM DESIGN

The performance of various gasifier and gasification system configurations as applied to the production and utilization of low and medium Btu gas was evaluated by examining the effect of gasification parameters on thermal efficiency for a given coal. Subsequently, the effect of varying coal feed on thermal efficiency/performance was estimated based on consideration of key constituents in the coal, i.e., moisture, sulfur, oxygen, and ash.

Effect of Gasification Parameters on Thermal Efficiency

The gasification parameters affecting thermal efficiency are oxidizing medium (air versus oxygen), pressure, and number of gasifier stages. A gasification system, which utilizes relatively pure oxygen for partial combustion of the coal to supply heat for the endothermic steam-carbon gasification reaction, usually has a higher thermal efficiency than if air were the oxidant. For the pressure effect, as the operating pressure increases, the driving force for the exothermic hydrogen-carbon reaction reduces the amount of oxidation required, thereby increasing the heating value of the gas produced and increasing the thermal efficiency. A two-stage, entrained-bed gasifier can reduce thermal losses by gasifying char produced in the low temperature stage (about 1800° F) in a high temperature stage. The gas from the high temperature stage

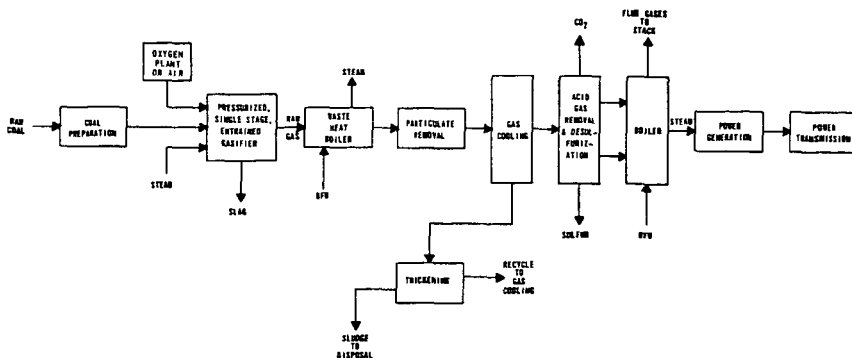


FIGURE 1
INTEGRATED GASIFICATION/CONVENTIONAL CYCLE
POWER PLANT SYSTEM

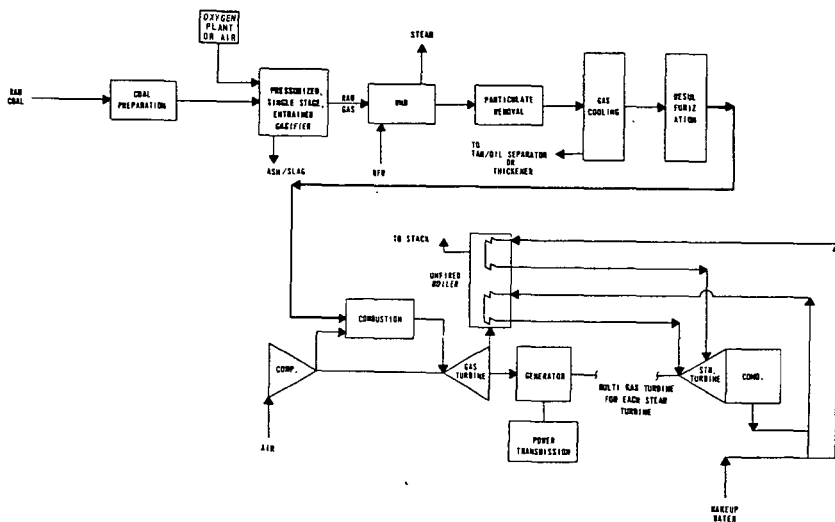


FIGURE 2
INTEGRATED GASIFICATION/COMBINED CYCLE
POWER PLANT SYSTEM

TABLE 3. TYPICAL FUEL CHARACTERISTICS FOR LOW AND MEDIUM-BTU GAS

	<u>Low-Btu Gas</u>		<u>Medium-Btu Gas</u>	
	<u>Low Pressure Entrained Bed</u>	<u>Low Pressure Entrained Bed</u>	<u>Pressurized Entrained Bed</u>	
Clean Gas Composition (% Dry)				
CO	22.24	52.73	29.54	
H ₂	17.18	36.13	32.36	
CO ₂	7.02	10.04	21.67	
CH ₄	0.03	-	15.83	
N ₂	<u>53.53</u>	<u>1.10</u>	<u>0.60</u>	
Total	100.00	100.00	100.00	
HHV: Btu/LB	120-130	280-290	358	
Stoichiometry Combustion Air, Lb/LB Fuel	1.05	3.03	4.02	

provides the heat for the coal feed stage. The two-stage, entrained-bed gasifier thus avoids the high coal combustion requirement that a single-stage, entrained-bed gasifier has (2700° to 3300° F).

Performance data for the gasification systems considered for power generation are presented in Table 4. The tabulation represents a combination of published data and engineering judgement applied in accordance with the effective system parameters outlined above. The hot and cold gas efficiencies for low pressure, single-stage, oxygen-blown, entrained-bed gasifiers⁽³⁾ (Case 3) and the low pressure, two-stage, air-blown, entrained-bed gasifier⁽⁴⁾ (Case 4) were obtained from published data. The efficiencies for Cases 1 and 2 were determined by taking into account pressure effects, i.e., increase of the thermal efficiencies by 1% for high pressure operation.

For producing electricity, when gasifiers are integrated with either a conventional or combined cycle power plant, the net station system efficiency is higher than the cold low or medium Btu gas efficiency but lower than the hot gas efficiency. Auxiliary power produced in the power plant and sensible heat recovered during the gas cleanup can be used as a part of the gasification system energy requirement. In general, integrating a gasification system with a power plant, will improve the efficiency of heat recovery and provide opportunities to optimize the overall cycle.

Integration of gasifiers with the combined cycle plant provides higher gasifier system efficiency than those with conventional power plants because of increased potential for cycle optimization. Additionally, for integration with the same power plant configuration, medium Btu gas provides a higher gasifier system efficiency than low Btu gas.

Table 4. Thermal Efficiency Of Gasification Systems

	Med.-Btu Integ. w/Base C.C.	Low-Btu Integ. w/Base C.C.	Med.-Btu Integ. w/Conv. Base	Low-Btu Integ. w/Conv. Base
Case	1	2	3	4
Type Gasifier	Pressurized Entrained Single Stage	Pressurized Entrained Two Stage	L.P. Entrained Single Stage	L.P. Entrained Two Stage
Oxidant	O ₂	Air	O ₂	Air
Coal Type	Ill. 6 Bit.	Ky. Bit.	Ill. 6 Bit.	Ky. Bit.
Gasifier Eff.				
Hot Gas, % (a)	92	93	91	92
Cold Gas, % (b)	76	77	75	76
Gasif. System Eff.%(c)	84.0	81.5	80.0	77.5
Power Plant Eff.%(c)				
Conventional Cycle	N/A	N/A	36.0	36.0
Combined Cycle	38.5	38.5	N/A	N/A
Integrated Gasifier/ Power Plant Eff., %(d)	32.3	31.4	28.8	27.9

(a) Hot gas efficiency,

$$\% = \frac{\text{HHV of gas @ gasifier exit temp.} + \text{sensible heat @ gasifier exit temp.}}{\text{HHV of coal fed to gasifier}} \times 100.$$

(b) Cold gas efficiency,

$$\% = \frac{\text{HHV of gas (after tar, oil, NH}_3\text{, H}_2\text{S have been removed)}}{\text{HHV of coal fed to gasifier}} \times 100.$$

(c) GAI estimate.

(d) Product of gasifier system efficiency and power plant efficiency.

In determining overall plant efficiencies for all the integrated cases, power plant efficiencies of 36.0% and 38.5% were used for conventional and combined cycle power plants, respectively.

Effect of Coal Feed on Thermal Efficiency

In order to facilitate an economic evaluation of alternatives, it was necessary to determine the effect of coal feed variation on the thermal efficiencies of the gasification systems and overall plants.

The key constituents of coal, which were considered in estimating the thermal efficiencies of a given process when fed with alternative coals, are moisture, sulfur, oxygen and ash.

- a. Moisture - Coal must be dry to about 3% moisture. The effect of moisture on gasifier system efficiency was determined by using a heat requirement of 1,000 Btu per pound of moisture.
- b. Sulfur - The gasifier system efficiency increases with decreasing sulfur content of coal. The effect of sulfur on efficiency was estimated by using the heating value of elemental sulfur.
- c. Oxygen - Highly reactive coals can be gasified at relatively lower temperatures than coals of low oxygen content. The low gasifier temperature requires less carbon combustion and increases thermal efficiency.
- d. Ash - As the ash content of coal increases, the amount of energy required in the coal preparation section for the dryer and pulverizer increases. Additionally, the energy losses in the gasifier system also increase with increasing ash content because increased power is required to feed the coal and some sensible heat is lost with ash leaving the gasifier.

The overall effect on thermal efficiency of these coal constituents was established for each coal in the study as a variance from the efficiency of the base coal. Typical results for variations in the gasifier system efficiencies from the based coal are summarized in Table 5.

ECONOMICS OF INTEGRATED SYSTEMS

Fixed Capital and Operating Costs

The base, fixed capital costs for all four integrated cases were estimated by adjusting published data⁽⁵⁾ to establish compatibility between the performance as proposed in the reference and that required to produce a desired fuel. The base, fixed operation and maintenance labor cost was estimated from a Combustion Engineering study⁽⁴⁾. The published data was adjusted using a power factor on electric generation capacity from a Fluor-Utah study⁽⁶⁾. The estimated base, fixed capital and operating costs are summarized in Table 6.

After the fixed capital and operating costs of each gasification system for base coals were established, the costs of each system when fed with alternative coals were determined using the calculated coal fuel rates, regional factors, and the scale factors required to adjust each cost element to compensate for the alternate coal feed. The fixed capital and operating costs for all cases considered are tabulated in Table 7.

TABLE 5. GASIFICATION SYSTEM
EFFICIENCIES FOR ALTERNATIVE COALS

	<u>BASE COAL</u>	<u>ILLINOIS NO. 6</u>	<u>ROSEBUD</u>	<u>MIDDLE KITTANING</u>	<u>STOCKTON</u>	<u>WYODAK</u>
Medium-Btu Gas Integrated With Combined Cycle	84.0 (Illinois 6)	84.0	84.0	88.0	90.0	84.0
Low-Btu Gas Integrated With Combined Cycle	81.5 (Kentucky 9)	81.5	83.5	85.5	87.5	81.5
Medium-Btu Gas Integrated With Conventional Cycle	80.0 (Illinois 6)	80.0	82.0	84.0	86.0	80.0
Low-Btu Gas Integrated With Conventional Cycle	77.5 (Kentucky 9)	77.5	79.5	81.5	83.5	77.5

TABLE 6. Summary of Low and Medium-Btu Gas Performance and Cost Data For Base Cases
Base Year: January - 1977

Case	1	2	3	4
Gasifier Type Selected	Medium-Btu Integrated w/Base C.C.	Low-Btu Integrated w/Base C.C.	Medium-Btu Integrated w/Conv. Base	Low-Btu Integrated w/Conv. Base
Region	MAIN	ECAR	MAIN	ECAR
Plant Capacity				
Net MWe	800	800	800	800
Gross MWe	810	810	827	831
Coal Type	Ill. 6 Bit.	Ky. Bit.	Ill. 6 Bit.	Ky. Bit.
As Received %				
Moisture	9.82	10.0	9.82	10.0
Carbon	64.16	63.4	64.16	63.4
Hydrogen	4.25	4.3	4.25	4.3
Nitrogen	1.42	1.4	1.42	1.4
Sulfur	3.35	3.2	3.35	3.2
Oxygen	5.36	6.8	5.36	6.8
Ash	11.64	10.9	11.64	10.9
Heating Value, Btu/Lb	11,390	11,400	11,390	11,400
Efficiencies, %				
Hot Gas(a)	92.0	93.0	91.0	92.0
Cold Gas(b)	76.0	77.0	75.0	76.0
Gasification System(c)	84.0	81.5	80.0	77.5
Overall Plant(d)	32.3	31.4	28.8	27.9
Coal Throughput, TPD	8,906	9,153	9,988	10,301
Fixed Capital Cost: MM\$				
Coal Preparation	21.72	22.15	23.53	24.04
Gasifier + Heat Recovery	54.01	73.26	70.22	95.43
Gas Purification	46.60	77.22	60.64	100.66
Sulfur Recovery	2.69	2.70	3.00	3.00
Oxygen Plant	36.34	-	39.38	-
Gasification Facility	33.72	30.35	36.54	32.88

TABLE 6. (Continued)

	Medium-Btu Integrated w/Base C.C.	Low-Btu Integrated w/Base C.C.	Medium-Btu Integrated w/Conv. Base	Low-Btu Integrated w/Conv. Base
Gasification Utility	50.60	45.55	54.83	49.35
Power Plant	227.40	236.41	277.60	291.80
Liquid Storage (Redundancy)	5.41	5.41	5.41	5.41
Gasif. Plant Land + Site Imp.	3.10	4.21	4.04	5.47
Total Fixed Capital Cost	481.61	497.26	575.19	608.04
Fixed Operating Cost: MM\$/Yr.				
Utility & Material	10.79	11.46	12.82	13.55
O&M Labor	3.83	3.83	3.40	3.40
General Overhead	7.73	7.84	9.18	9.70
Total Fixed Operating Cost	22.35	23.13	25.40	26.65

86 (a) Hot Gas Eff. % = $\frac{(\text{HHV of gas} + \text{sensible heat}) @ \text{gasifier exit temp.}}{\text{HHV of coal fed to gasifier}}$

(b) Cold Gas Eff. % = $\frac{\text{HHV of gas (after H}_2\text{S, COS, NH}_3\text{, tar removal)}}{\text{HHV of coal fed to gasifier}}$

(c) GAI estimate (see Table 5).

(d) Multiplication of gasification system efficiency with power plant efficiencies of 38.5% for combined cycle plant and 36.0% for conventional plant (see Table 5).

TABLE 7. SUMMARY OF FIXED CAPITAL AND OPERATING COSTS FOR ALTERNATE COALS

BASIS: JANUARY, 1977 PRICING

FIXED CAPITAL COST: MM\$	MAIN				NPCC			
	ILLINOIS		HIGH SULFUR COAL		MIDDLE		HIGH SULFUR COAL	
	NO. 6		ROSEBUD		KITTANING		ROSEBUD	
	STOCKTON		WYODAK		STOCKTON		WYODAK	
MED-BTU W/COMB. CYCLE	479.6	493.4	418.9	469.4	461.0	498.3	423.1	474.1
LOW-BTU W/COMB. CYCLE	468.6	479.2	387.3	433.8	450.2	484.2	391.3	438.3
MED-BTU W/CONV. CYCLE	574.1	589.3	499.8	555.8	55.28	595.2	504.8	561.4
LOW-BTU W/CONV. CYCLE	573.8	588.8	472.2	526.6	551.6	594.7	476.9	531.6
FIXED OPER. COST: MM\$/YR								
MED-BTU W/COMB. CYCLE	22.3	22.6	19.8	21.9	21.6	22.8	20.1	22.1
LOW-BTU W/COMB. CYCLE	22.2	22.5	18.8	20.7	21.5	22.8	19.0	20.9
MED-BTU W/CONV. CYCLE	25.3	26.1	22.3	24.6	24.4	26.4	22.5	24.8
LOW-BTU W/CONV. CYCLE	26.3	27.1	22.1	24.3	25.4	27.4	22.3	24.6

TABLE 8. Financial Parameters Used to
Develop Power Generation Cost

Plant Life	20 Years
Depreciation (Based on Total Capital Less Working Capital)	5%/Year Straight Line
Fraction Debt	0.75
Return on Equity	15%/Year
Interest on Debt	12%/Year
Load Factor	70%
Working Capital	Coal Inventory for 60 Days and 1% of Fixed Capital Cost
Interest During Construction	Interest on Debt x Total Fixed Capital x 2
Federal Income Tax Rate	48%

Development of Power Generation Cost

The fixed capital and operating costs summarized in Tables 2 and 7 were used to develop power generation cost; the utility financing method was used with the financial parameters summarized in Table 8. The power generation costs calculated are summarized in Table 9 together with the delivered coal cost.

CONCLUSIONS

Referring to Table 9, the following conclusions were observed:

1. The western coals (both high and low sulfur) in the load center sites of the NPCC region are not competitive with eastern coals, whereas the western coals are competitive with the eastern coals in the MAIN region.
2. In the MAIN region, both eastern and western high sulfur coals are competitive with low sulfur coals. In the NPCC region, however, the eastern high sulfur coal appears to be more attractive than the eastern low sulfur coals.
3. Integrated conventional plants in both regions for all coals are not competitive with the two conventional power plants using high sulfur coal with FGD and low sulfur without FGD.
4. Integrated combined cycle plants using the eastern high sulfur coal in the NPCC region are more attractive than the two conventional power plants.
5. In the MAIN region, the integrated combined cycle plants are either better than or comparable to the high sulfur coal fired plants with FGD, whereas they are not competitive with the low sulfur coal fired plants without FGD.

TABLE 2. SUMMARY OF POWER GENERATION COST

BASIS: JAN-1977 ¢/KWH

NERC REGION HIGH OR LOW SULFUR COAL BED	MAIN				NPCC			
	HIGH SULF. ILL. 6	ROSEB.	STOCKT.	LOW SULF. WYOD.	HIGH SULF. M-KIT.	ROSEB.	STOCKT.	LOW SULF. WYOD.
COAL COST, \$/TON DELIVERED FOB MINE	24.81 19.55	20.95 9.22	39.48 31.97	18.92 7.19	27.37 21.08	27.42 9.22	39.13 31.97	25.38 7.19
LOW SULFUR COAL WITHOUT FGD	-	-	3.065	2.751	-	-	3.067	3.171
HIGH SULFUR COAL WITH FGD	3.340	3.276	-	-	3.316	3.616	-	-
MED-BTU GAS INT. W/COMB. CYCLE	3.202	3.163	3.310	3.256	3.031	3.512	3.317	3.702
LOW-BTU GAS INT. W/COMB. CYCLE	3.197	3.147	3.230	3.149	3.026	3.508	3.233	3.606
MED-BTU GAS INT. W/CONV. CYCLE	3.718	3.684	3.811	3.751	3.524	4.077	3.817	4.251
LOW-BTU GAS INT. W/CONV. CYCLE	3.779	3.740	3.766	3.691	3.577	4.145	3.770	4.208

All observations were based on the fixed delivered coal cost. In order to determine coal cost situations where the integrated combined cycle plants in the MAIN region would be competitive with low sulfur conventional coal fired plants without FGD, the sensitivity of the integrated of the plant generation cost to coal cost is analyzed, as shown in Figure 3. The lowest power generation cost in the main region was 2.751¢ per kilowatt hour for Wyodak coal without FGD. Figure 3 indicates that for medium Btu gas integrated with a combined cycle power plant, the delivered coal prices would have to be \$15.00, \$12.50 & \$25.00 per ton of Illinois No. 6, Rosebud, and Stockton coals respectively to be competitive with the Wyodak coal fired without FGD. The study was intended solely to demonstrate how the selection of coal feedstocks and regions effect the power generation costs for various configurations.

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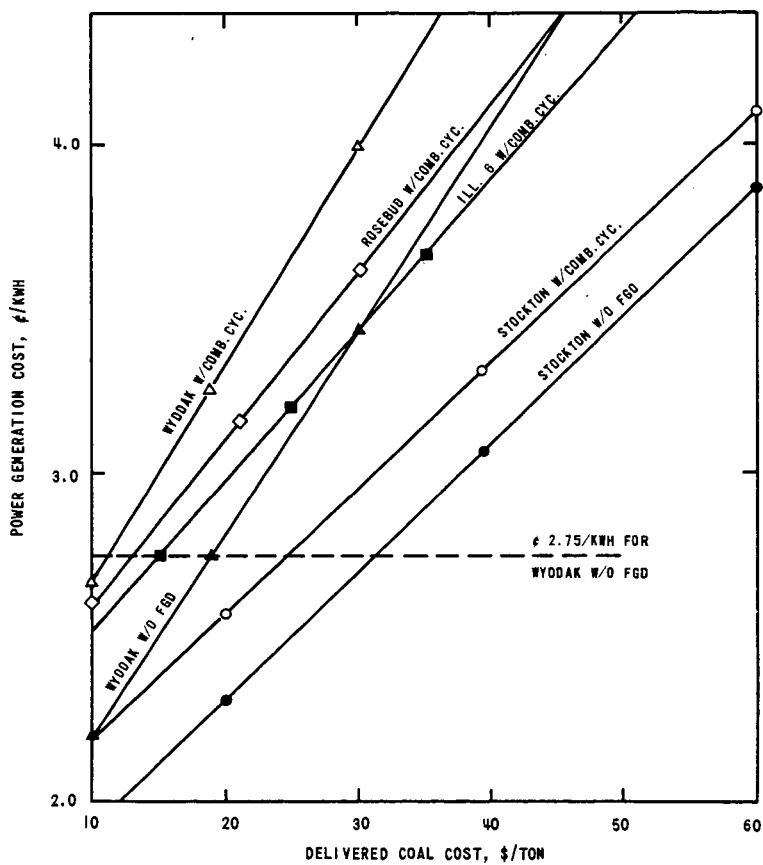


FIGURE 3
SENSITIVITY OF POWER GENERATION
COST TO DELIVERED COAL COST

HIGH- AND LOW-Btu GAS FROM MONTANA
SUBBITUMINOUS COAL

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INTRODUCTION

Two coal gasification processes are under development at IGT. The HYGAS[®] Process has been developed for high-Btu gas (SNG) from coal; the U-GAS[®] Process, a much simpler system, has been developed for low-Btu gas. This paper describes the application of these gasifiers for different objectives and compares process and economic characteristics. HYGAS and U-GAS reactor systems are compared for the manufacture of pipeline gas, and the U-GAS Process is analyzed as an advantageous source of low-Btu gas. Three process designs and their economics for manufacturing a nominal amount of 240 billion Btu/day of product gas are discussed. The designs are based on the conversion of Montana subbituminous coal, whose analysis is given in Table 1. Because the coal is nonagglomerating, pretreatment is not required.

Table 1. MONTANA SUBBITUMINOUS COAL

<u>Proximate Analysis</u>	<u>Weight Percent</u>
Moisture	22.0
Volatile Matter	29.4
Fixed Carbon	42.6
Ash	6.0
Total	100.0
<u>Ultimate Analysis (Dry)</u>	
Carbon	67.70
Hydrogen	4.61
Nitrogen	0.85
Oxygen	18.46
Sulfur	0.66
Ash	7.72
Total	100.00
Dry Heating Value, Btu/lb	11,290

PROCESS DESIGNS FOR PIPELINE GAS (HIGH-Btu GAS)

Two process designs for the manufacture of 242 billion Btu/day of SNG at 1000 psig from coal have been made: one based on the HYGAS Process and a similar design utilizing the U-GAS Process. The capacity was set by an existing design based on the HYGAS Process. A comparison of the two processes will show any economic benefit derived from the use of the more complex and costly HYGAS reactor in contrast to the simpler U-GAS reactor in the manufacture of pipeline gas from coal.

Comparison of the HYGAS and U-GAS Reactors

The HYGAS reactor (hydrogasifier) is designed to maximize direct methane formation by the reaction



This reaction supplies heat for the endothermic reaction also occurring in the hydrogasifier:



High pressure in the reactor, 1165 psig in this design, favors the formation of methane.

Process coal at a rate of 15,996 tons/day is dried to 10% moisture and simultaneously ground to below 8 mesh with a maximum of 15% below 100 mesh. The prepared coal is pneumatically conveyed to the slurry preparation section, and a 50% water slurry is pumped to the hydrogasifier. A fluidized-bed dryer is located at the top of the vessel, where the slurry water is vaporized in contact with the hot reactor effluent gases.

The reactor coal feed passes through three zones of conversion: 1) a low-temperature (1000°F) transport reactor, where the coal is devolatilized and rapid-rate conversion to methane enriches the product gas; 2) the main fluidized bed at 1700°F, where most of the methane is formed; and 3) the steam-oxygen gasification zone at 1850°F, where synthesis gas is generated from the hydrogasifier char according to the endothermic steam decomposition reaction



Heat is supplied by partial combustion of the char with oxygen:



Further generation of hydrogen occurs in zone 2, where the exothermic methane formation reaction supplies heat for the steam decomposition reaction (Reactions 1 and 2).

The U-GAS reactor is a single-stage fluidized-bed gasifier operating at 1900°F and 335 psig. The reactor is not primarily designed to make methane. To promote methane formation, where SNG is the desired end product, 18,400 tons/day of coal is fed into the upper portion of the gasifier onto the fluidized bed. The countercurrent flow of hot gases and coal devolatilizes the coal, and some methane is formed. Reactions 2 and 4 are the major reactions taking place in this system. A lockhopper coal feed system, which is used commercially at this relatively low pressure level, is used to feed the coal. Further operating details of the U-GAS system are discussed in the section on low-Btu gas.

Raw gas compositions from the two reactors are compared in Table 2. The total moles per hour is the requirement for 242 billion Btu/day of product gas.

Table 2. COMPOSITION OF RAW GAS FROM GASIFIERS

	HYGAS Hydrogasifier Effluent	U-GAS Raw Gas
	mol %	
CO	20.13	34.18
CO ₂	18.65	13.30
H ₂	23.68	29.52
H ₂ O	22.68	17.44
CH ₄	12.86	4.84
C ₂ H ₆	0.99	--
NH ₃	0.34	--
H ₂ S	0.19	0.20
N ₂ + Ar	0.18	0.52
B-T-X	0.30	--
	100.00	100.00
Total mol/hr	103,288	126,576

In addition to coal raw material, generation of these gases requires steam and oxygen. The HYGAS reactor requires 1,003,130 lb/hr of steam at 1200 psig and 1050°F, plus 2999 tons/day of 98% oxygen. The U-GAS reactor requires 670,320 lb/hr of steam at 385 psig and 800°F, plus 7986 tons/day of oxygen.

The Manufacture of Pipeline Gas

The raw gases from both reactors require upgrading to pipeline-gas quality. For the HYGAS plant, the required steps are shown in the flow diagram of Figure 1, and the compositions of the process flow streams are given in Table 3. Figure 2 and Table 4 give similar information for the U-GAS plant.

SNG by HYGAS

The effluent gas is cooled by waste heat recovery and cleaned in a venturi scrubber to remove small particles carried over from the hydrogasifier. The gas is sent to a CO conversion reactor where the H₂/CO ratio is raised to 3.2 or 3.3 in preparation for methanation. The catalyst is an oil- and sulfur-resistant, high-temperature CO conversion catalyst. Steam for this reaction is supplied by vaporized slurry feedwater present in the raw gas.

The B-T-X formed in the hydrogasifier is recovered as a valuable by-product after CO conversion. Oil scrubbing and activated carbon are used for this operation. Large amounts of CO₂ and H₂S must be removed from the gas during the upgrading to pipeline gas quality. This is done by hot carbonate scrubbing; acid gases leaving this section are sent to a Stretford unit for sulfur recovery. Final traces of H₂S are removed by activated carbon and zinc oxide beds.

Table 3. PROCESS FLOW STREAMS FOR A NOMINAL 240 X 10⁹ Btu/DAY HIGH-Btu GAS PLANT BY THE HYGAS PROCESS FROM MONTANA SUBBITUMINOUS COAL

Stream Name	Hydrogasifier Effluent		Slurry Water Vaporizer Effluent		CO Shift Feed		B-T-X Recovery Feed		Hot K ₂ CO ₃ Feed		Methanation Feed		Pipeline Gas	
	1	2	2	3	4	5	6	7	8	9	10	11	12	13
Stream No.	1000	600	1155	483	125	125	125	100	100	100	100	100	100	100
Temperature, °F	1165	1155	1155	1145	1110	1100	1075	1075	1075	1075	1075	1075	1075	1075
Pressure, psig	1165	1155	1155	1145	1110	1100	1075	1075	1075	1075	1075	1075	1075	1075
Components														
CO	20.13	20.793	12.91	20.793	13.03	20.793	12.03	10.514	12.06	10.505	17.45	10.477	0.10	28
CO ₂	18.65	19.264	11.97	19.264	12.07	19.264	31.36	27.393	31.30	27.255	1.00	599	1.31	362
H ₂	23.68	24.455	15.19	24.455	15.32	24.455	39.70	34.678	39.82	34.678	57.46	34.504	4.37	1,207
H ₂ O	22.68	23.429	50.40	81.140	49.95	79.704	0.17	150	0.17	150	0.18	107	0.01	4*
CH ₄	12.86	13.288	8.25	13.288	8.33	13.288	15.16	13.240	15.19	13.229	21.94	13.178	93.54	25,863
C ₂ H ₆	0.99	1.020	0.63	1.020	0.64	1.020	1.16	1.014	1.15	1.002	1.66	998	--	--
NH ₃	0.34	348	0.22	348	0.22	348	--	--	--	--	--	--	--	--
B-T-X	0.30	305	0.19	305	0.19	305	0.13	114	0.02	16	--	--	--	--
H ₂ S	0.19	201	0.12	201	0.13	201	0.08	71	0.08	67	--	--	--	--
N ₂ + Ar	0.18	185	0.12	185	0.12	185	0.21	185	0.21	185	0.31	185	0.67	185
Total	100.00	103.288	100.00	160.999	100.00	159.563	100.00	87.359	100.00	87.087	100.00	60.048	100.00	27,649

* 7 lb H₂O/10⁶ SCF.

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Table 4. PROCESS FLOW STREAMS FOR A NOMINAL 240×10^9 Btu/DAY HIGH-Btu GAS PLANT BY THE U-GAS PROCESS FROM MONTANA SUBBITUMINOUS COAL

Stream Description	Raw Gasifier Product		CO Shift Feed		Syn Gas Compressor Feed		Methanation Feed		Pipeline Gas	
	mol %	mol/hr	mol %	mol/hr	mol %	mol/hr	mol %	mol/hr	mol %	mol/hr
Stream No.	1		2		3		4		5	
Temperature, °F	1700		376		100		25		128	
Pressure, psia	350		350		285		440		1015	
	335		335		270		425		1000	
Component										
CO	34.18	43,262	19.41	43,262	14.88	19,199	21.66	19,175	0.10	29
CO ₂	13.30	16,832	7.55	16,832	31.71	40,895	1.00	885	1.13	324
H ₂	29.52	37,373	16.77	37,373	47.63	61,436	69.26	61,318	5.71	1,624
H ₂ O	17.44	22,081	53.11	118,364	0.33	429	0.43	382	0.01	4
CH ₄	4.84	6,121	2.75	6,121	4.75	6,121	6.90	6,113	90.74	25,823
H ₂ S	0.20	248	0.11	248	0.19	248	0.1 ppmv	--	--	--
N ₂ + Ar	0.52	659	0.30	659	0.51	659	0.75	659	2.31	659
Total	100.00	126,576	100.00	222,859	100.00	128,987	100.00	88,532	100.00	28,460

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The purified gas is methanated in a fixed-bed reactor where essentially all the CO and some of the CO₂ are converted by the following reactions:



Temperature is controlled by recycling the product so as to dilute the CO content in the feed mixtures to the four reactor stages to about 4%. This limits the maximum catalyst bed temperature to 900°F. A product gas of 961 Btu/SCF HHV leaves the plant at 1000 psig.

Water condensate from CO conversion effluent goes through oil-water separation and a Chevron waste-water treatment process. Stripped gases go to an ammonia recovery section where 69 short tons/day are recovered as by-product. Acid gases are combined with those from the hot carbonate section and sent to the Stretford unit. The by-product sulfur is 65.3 long tons/day. Total by-product B-T-X recovery is 84,144 gal/day.

SNG by U-GAS

The flow diagram for this process (Figure 2) shows major steps similar to those for the HYGAS Process. However, there are several important differences.

1. Because of the much lower operating pressure, the U-GAS system uses lockhoppers to feed the dried, ground coal to the reactor instead of slurry feed.
2. We have assumed that ammonia is not formed, and since the U-GAS reactor does not make B-T-X, recovery systems for these materials are not required.
3. The steam for CO conversion is generated by adiabatic humidification of the hot (1700°F) raw gas in the venturi scrubber, recovering heat in cooling to 380°F.
4. Because of the lower gasifier pressure compared with HYGAS (335 vs. 1165 psig), subsequent compression to 450 psig before acid-gas removal and final product compression to 1000 psig are required.

Comparison of HYGAS and U-GAS Processes for the Manufacture of Pipeline Gas

Gasifier and process parameters, process energy balances, and efficiencies for the manufacture of pipeline-quality gas by the HYGAS and U-GAS Processes are shown in Tables 5, 6, and 7. The utility requirements for each process design were estimated, and complete energy balances were made. Both plants have coal-fired boilers for steam and power generation.

The gasifier feed quantities are presented in Table 5. The U-GAS reactor consumes about 15% more coal than the HYGAS reactor at equal carbon conversions of 98%. However, the steam requirement for U-GAS is about 67% of that for HYGAS; this is because the U-GAS reactor operates at 1900°F and HYGAS has reaction zones at 1000°, 1700°, and 1850°F, so the reaction rates are higher. The most significant difference in gasifier feeds is in the amount of oxygen. The U-GAS reactor requires 7986 tons/day of oxygen, which is about 2.7 times as much as required by the HYGAS reactor. The proportionately larger U-GAS oxygen plant is one of the major factors contributing to the greater utility requirements and higher costs for U-GAS as compared with HYGAS.

Table 6 is a comparison of important process quantities for each design. The HYGAS reactor operates at over 1000 psig as compared with the 335 psig operating

Table 5. COAL, REACTOR STEAM, AND OXYGEN REQUIREMENTS FOR MANUFACTURING
NOMINAL 240 X 10⁹ Btu/DAY HIGH- AND LOW-Btu GAS FROM MONTANA SUBBITUMINOUS COAL

	High-Btu Gas ^a		Low-Btu Gas	
	HYGAS	U-GAS	U-GAS	
Reactor Coal, lb/hr (dry)	1,039,728	1,199,943	987,553	
Fuel Coal, lb/hr (dry)	232,974	335,931	105,682	
Total Coal	1,272,702	1,535,874	1,093,235	
Total Coal (22% moisture), tons/day	19,580	23,629	16,819	
Reactor Steam, lb/hr	1,003,131	670,321	551,724	
Oxygen, tons/day (98% purity)	2,999	7,986	6,573	
CO-Shift Steam, lb/hr*	957,298 (Supplied by slurry vaporizer)	1,421,625 (Supplied by injecting BFW to cool U-GAS effl.)	--	

* Steam going to the CO shift reactor (2/3 of total feed).

Table 6. COMPARISON OF PROCESS QUANTITIES FOR MANUFACTURING
NOMINAL 240 X 10⁹ Btu/DAY HIGH- AND LOW-Btu GAS
FROM MONTANA SUBBITUMINOUS COAL

	High-Btu Gas		Low-Btu Gas
	HYGAS	U-GAS	U-GAS
Gasifier Pressure, psig	1,165	335	335
Gasifier Temperature, °F	1,000-1,850	1,700-1,900	1,700-1,900
CH ₄ in Gasifier Effluent, mol/hr	13,288	6,121	5,038
Percent of Product Methane Made in Gasifier	51	24	100*
C ₂ H ₆ in Gasifier Effluent, mol/hr	1,020	--	--
CO + H ₂ in Gasifier Effluent, mol/hr	45,248	80,635	66,363
CO Shifted, mol/hr	10,246	24,063	--
CO ₂ + H ₂ S + COS Removal, mol/hr	26,723	40,258	3,477
CH ₄ Made in Methanator, mol/hr	12,685	19,710	--
Total CH ₄ in Product Gas, mol/hr	25,863	25,823	4,931
Plant Power Required, kW	101,814	238,172	138,107
Plant Electric Motors, kW	46,602	52,523	53,196
Plant Steam or Expansion Turbine Drives, equivalent kW	55,212	185,649	4,374 [†]
Process Cooling Water, gpm	36,610	82,662	44,572
Turbine Driver Condenser Cooling Water, gpm	29,090	113,305	--
Power Plant Cooling Water, gpm	31,810	31,265	31,225
Plant Raw Water Required, gpm	4,275	8,223	3,115
Product Gas Heating Value, 10 ⁹ Btu/day	241.5	242.3	238.8
Product Gas Heating Value, Btu/SCF	961	937	320

* Methanation unnecessary for low-Btu gas.

† Expansion turbine.

Table 7. OVERALL ENERGY BALANCES AND PROCESS EFFICIENCIES FOR MANUFACTURING
NOMINAL 240 X 10⁶ Btu/DAY HIGH- AND LOW-Btu GAS FROM MONTANA SUBBITUMINOUS COAL

	High-Btu Gas		Low-Btu Gas	
	HYGAS	U-GAS	U-GAS	U-GAS
	10 ⁶ Btu/hr			
HHV Total Coal Input	14,368.8	17,340.0	12,342.3	
Product Gas, HHV	10,061.6	10,097.6	9,953.1	
	Output, % of Coal HHV			
Product Gas	70.0	58.2	80.6	
By-product Chemicals	4.0	0.2	0.2	
Process Vent Gases	3.0	3.0	3.1	
Stack Gases	2.1	2.9	1.0	
Heat Dissipated to Cooling Water and Air	17.2	31.8	10.6	
Gasifier Residue	1.5	1.5	1.7	
Assumed and Unaccounted Losses	2.2	2.4	2.8	
Total	100.0	100.0	100.0	
Overall Plant Efficiency, Coal to Products	74.0	58.4	80.8	

pressure for the U-GAS reactor. Because of the higher operating pressure and the multistage hydrogasification reaction, HYGAS produces more methane in the reactor: 13,288 mol/hr of CH_4 and 1,020 mol/hr of ethane as compared with 6,121 mol/hr of methane for U-GAS.⁴ The amount of methane in the product gas is about the same (25,800 mol/hr) for both designs. However, the U-GAS reactor makes only 24% of this total as compared with 51% by the HYGAS reactor. To achieve the same total plant output of methane, a U-GAS system requires more synthesis gas, hence more oxygen, and bigger CO shift, acid-gas removal, and methanation sections. The comparable quantities of CO shifted, acid-gas removed, and methane made in the methanator for both the HYGAS and U-GAS designs are shown in Table 6.

Table 6 also indicates the substantially higher power, cooling water, and raw water requirements for the U-GAS design due to the higher oxygen usage and to the power requirement for product gas compression to 1000 psig.

Table 7 presents a comparison of overall energy balances and process efficiencies. SNG via the U-GAS Process requires about 20% more plant coal, and the coal-to-pipeline gas efficiency is 58.2% versus 70% for the HYGAS system. In addition, HYGAS has 4.0% of the feed coal HHV converted to by-products, whereas the U-GAS system has only 0.2% converted, raising the HYGAS plant efficiency. The U-GAS system has considerably more heat dissipated to cooling media: 5504 vs. 2471 million Btu/hr, or 31.8% vs. 17.2% of plant coal feed. The HYGAS system heat loss to cooling water is less than half that for the U-GAS system. This is primarily due to the very large difference in the amount of cooling necessary for the condensers on the plant turbine drivers, 84,215 gpm. The difference in process cooling, while significant, is relatively minor by comparison. Overall efficiencies (coal to all products) are 74.0% for HYGAS and 58.4% for U-GAS.

LOW-Btu GAS BY THE U-GAS PROCESS

Figure 3 shows the flow diagram for producing low-Btu gas by the U-GAS Process, an appropriate application for this process, and the process flow streams are given in Table 8. The results are more favorable than in the SNG application and are shown in Tables 5, 6, and 7. To put this plant on a comparable basis with the other plants in this study, the same product fuel value output rate was used for all three. When making low-Btu instead of high-Btu gas with the U-GAS reactor, the process coal feed is reduced to 15,193 tons/day of Montana subbituminous coal, and the plant produces 239 billion Btu/day of 320 Btu/SCF fuel gas.

For the low-Btu U-GAS reactor process, coal is dried to 10% moisture and ground to 1/4 in. X 0. Lockhoppers introduce the coal to the gasifier. Simultaneous with gasification, ash is removed from the fluidized bed by an ash-agglomerating technique, and fines elutriated from the bed returned through cyclones. The gasifier requires 551,724 lb/hr of steam and 6,573 tons/day of oxygen. Raw gas is cooled to 315°F in a waste heat boiler and is water-scrubbed in a venturi scrubber for dust removal.

Some adiabatic humidification occurs in the scrubber that cools the gas to 293°F. Prior to H_2S removal, the gas is cooled to 100°F, and the condensed water is sent to waste-water treating facilities and used as cooling tower makeup.

The hydrogen sulfide in the raw gas is removed by the Selexol Process. Besides hydrogen sulfide, a small amount of carbonyl sulfide is produced in the gasifier, and this compound is also partly removed by the Selexol Process. The total sulfur present in the clean gas is reduced to about 70 ppm. Together with hydrogen sulfide, the process removes about 24% of the carbon dioxide present in the raw gas. The H_2S - CO_2 mixture from the Selexol unit is sent to a Streford unit where 68 long tons/day of sulfur is recovered. The clean desulfurized gas from the Selexol absorber

Table 8. PROCESS FLOW STREAMS FOR NOMINAL 240 X 10⁹ Btu/DAY LOW-Btu GAS
BY THE U-GAS PROCESS FROM MONTANA SUBBITUMINOUS COAL

Stream Name	Raw Gas		Scrubber Effluent		Selsol Feed		Selsol Effluent to Expander		H ₂ S Stream to Sulfur Recovery Unit (Streibord)		Product Gas	
Stream No.	1		2		3		4		5		6	
Temperature, °F	1700		293		100		85		115		100	
Pressure, psig	335		320		310		300		10		8	
Component	mol/hr	mol %	mol/hr	mol %	mol/hr	mol %	mol/hr	mol %	mol/hr	mol %	mol/hr	mol %
CO	35,605	34.17	35,605	33.90	35,605	41.29	35,306	43.06	299	7.04	35,306	43.06
CO ₂	13,843	13.29	13,843	13.18	13,843	16.05	10,564	12.88	3,279	77.17	10,564	12.88
H ₂	30,758	29.53	30,758	29.29	30,758	35.67	30,639	37.37	119	2.80	30,369	37.37
CH ₄	5,038	4.84	5,038	4.80	5,038	5.84	4,931	6.01	107	2.52	4,931	6.01
H ₂ S	194	0.19	194	0.18	194	0.22	1	--	193	4.54	173	2.13
COS	10	0.01	10	0.01	10	0.01	5	0.01	5	0.12	5	0.12
N ₂ + Ar	542	0.52	542	0.52	542	0.63	539	0.66	3	0.07	539	0.66
H ₂ O	18,183	17.45	19,037	18.12	252	0.29	8	0.01	224	5.74	8	0.01
Total	104,173	100.00	105,027	100.00	86,242	100.00	81,993	100.00	4,249	100.00	81,993	100.00

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is heated to 700°F and then expanded in a power recovery turbine. Most of this energy (108,000 hp) is used to drive the oxygen plant air compressors, which are coupled to the expander shaft; also, 5,866 kW of electricity is generated and used to drive plant motors. The expanded gas is cooled to 100°F and sent to boilers.

COMPARISON OF HIGH-Btu (HYGAS) AND LOW-Btu (U-GAS) PROCESSES

Both the HYGAS and the U-GAS Processes provide alternative energy sources through coal conversion techniques. The process differences result because each is specifically designed for the form of energy product desired. The SNG from HYGAS is for the higher valued pipeline gas, while the low-Btu gas from U-GAS is designed for use as industrial boiler fuel for process steam generation or for combined gas turbine-steam turbine power cycles.

The U-GAS system is simpler than the HYGAS system because it requires no equipment to produce methane or remove liquid hydrocarbons. For example, the U-GAS Process does not require CO conversion, benzene recovery, methanation, or CO₂ removal (the HYGAS Process uses the hot carbonate system, which removes CO₂, and the U-GAS Process uses Selexol, which minimizes CO₂ removal).

The gasifier inputs, process quantities, energy balances, and process efficiencies are presented in Tables 5, 6, and 7 for the high- and low-Btu gas processes. Both plants have boilers for steam and/or power generation.

Table 5 presents coal, gasifier steam, and oxygen requirements. The HYGAS reactor requires 5% more coal than the U-GAS reactor, but the total coal needed, including fuel coal, is 16% more for the HYGAS Process. Fuel coal for U-GAS is less than half that for HYGAS because of the large amount of power recovered by expanding the product gas down to 10 psig. The U-GAS oxygen requirement is 6,573 tons/day, which is over twice the HYGAS requirement. This disadvantage in oxygen plant costs and utilities is more than compensated for by the much simpler product upgrading when making low-Btu gas. The HYGAS reactor requires 80% more steam than the U-GAS reactor, and HYGAS also requires about 960,000 lb/hr of CO-shift steam.

In Table 6 process quantities for the two processes are compared. Plant power required is about 38% more for the U-GAS system because of the larger oxygen plant. The HYGAS total cooling water requirement is about 29% more than for U-GAS and the raw water requirement for HYGAS is 37% more than for U-GAS. Acid-gas removal for HYGAS is 26,723 mol/hr and only 3,477 mol/hr for U-GAS. The overall plant efficiency for low-Btu gas is 80.8% compared with 74% for the high-Btu gas (HYGAS) (Table 7).

COMPARISON OF PROCESS ECONOMICS FOR COAL TO HIGH- AND LOW-Btu GAS USING HYGAS AND U-GAS PROCESSES

Capital and annual operating costs for high- and low-Btu gas processes are estimated on a comparable basis in mid-1976 dollars and are given in Tables 9 and 10. These costs do not include stack-gas cleanup because sulfur in the Montana coal is low enough to meet the emission specifications of 1.2 lb SO₂/million Btu of solid fuel burned. If the standards change in the future, stack-gas cleanup may be required. The annual operating costs and returns on investment are based on the utility financing method of the Supply-Technical Advisory Task Force - Synthetic Gas-Coal for the FPC National Gas Survey. The basic assumptions of this method are given in Table 11.

Table 9. CAPITAL INVESTMENT SUMMARY FOR NOMINAL
240 X 10⁹ Btu/DAY HIGH- AND LOW-Btu GAS FROM
MONTANA SUBBITUMINOUS COAL
(Mid-1976 Costs)

Section	High-Btu Gas		Low-Btu Gas
	HYGAS	U-GAS	U-GAS
	\$10 ⁶		
Coal Storage - Reclaiming	5.0	6.0	4.3
Coal Grinding and Drying	13.7	14.3	11.8
Coal-Water Slurry Feed System			
(Lock Hoppers for U-GAS)	11.1	4.0	3.3
Slurry Feed Preheat (Fired Heater)	4.5	--	--
Gasifiers	43.0	22.3	18.4
Char Residue and Plant Ash Disposal	2.4	2.8	2.3
Gasifier Effluent Dust Removal System	3.8	4.5	4.0
Carbon Monoxide Conversion	11.2	10.0	--
Benzene Recovery	5.6	--	--
Prepurification (Hot K ₂ CO ₃ , Bulk, Activated Carbon, Zinc Oxide -- Selexol for U-GAS Low-Btu Gas Case)	46.6	50.6	19.6
SYN Gas Compressors or Expander	--	12.0	13.7*
Methanation, Drying, and Product Gas Compression	15.2	37.3	--
Process Waste-Heat Recovery	14.2	5.1	15.7
High-Pressure Oxygen Supply	45.0	109.0	89.6
Process and Turbine Steam Generation	69.1	84.6	28.4
Turbogenerator	7.7	7.4	7.6
Electric Power Distribution	7.7	8.7	9.5
Cooling and Plant Makeup Water	4.9	8.5	3.1
Sulfur Recovery -- Stretford	16.0	19.0	16.8
Waste-Water Treatment	13.1	3.0	6.5
Particulate-Emission Control	3.8	4.5	3.2
Miscellaneous	17.2	20.7	12.9
General Facilities	43.3	47.7	32.5
Installed Plant Cost, Excluding Contingencies	404.1	482.0	303.2
Contingencies at 15%	60.6	72.3	45.5
Total Bare Cost	464.7	554.3	348.7
Contractor's Overhead and Profits (15%)	69.7	83.1	52.3
Total Plant Investment (I)	534.4	637.4	401.0
Interest During Construction (9% X 1.875 years X I)	90.2	107.6	67.7
Start-up Cost (5% of Total Plant Investment)	26.7	31.9	20.1
Working Capital: 60 days' coal at full rate	10.4	12.5	8.9
0.9% of Total Plant Investment	4.8	5.7	3.7
1/24 X Annual Revenue Required	7.3	9.4	6.1
Total Capital Required	673.8	804.5	507.5

* Expander.

Table 10. ANNUAL OPERATING COSTS FOR NOMINAL
240 X 10⁹ Btu/DAY HIGH- AND LOW-Btu GAS PLANTS
USING MONTANA SUBBITUMINOUS COAL
(90% Plant Service Factor - Timing: Mid-1976)

<u>Operating Cost Component</u>	<u>High-Btu Gas</u>		<u>Low-Btu Gas</u>
	<u>HYGAS</u>	<u>U-GAS</u>	<u>U-GAS</u>
		\$1000	
Coal Feed, 50¢/10 ⁶ Btu*	56,641	68,353	48,653
Catalysts, Chemicals and Other Direct Materials	3,195	5,236	847
Raw Water Cost, 45¢/1000 gal	909	1,749	670
<u>Labor</u>			
Process Operating Labor (for high-Btu gas, 58 men/shift for HYGAS and 60 men/shift for U-GAS; 33 men/shift for U-GAS to low-Btu gas; at \$7.20/hr and 8,760 man-hr/year)	3,659	3,784	2,081
Maintenance Labor (1.5% of Total Plant Investment plus Lockhopper Maintenance Labor for U-GAS)	8,016	9,661	6,115
Supervision (15% of Operating and Maintenance Labor)	1,751	2,017	1,229
Administration and General Overhead (60% of Total Labor, Including Supervision)	8,056	9,277	5,655
<u>Supplies</u>			
Operating (30% of Process Operating Labor)	1,098	1,135	624
Maintenance (1.5% of Total Plant Investment plus Lockhopper Maintenance Supplies for U-GAS)	8,016	9,661	6,115
Local Taxes and Insurance (2.7% of Total Plant Investment)	14,429	17,210	10,827
Total Gross Operating Cost	105,770	128,083	82,816
<u>By-Product Credits</u>			
Sulfur at \$10/long ton	(215)	(267)	(223)
Ammonia at \$50/ton	(1,138)	--	--
Light Oil (B-T-X) at 35¢/gal	(9,674)	--	--
Total	(11,027)	(267)	(223)
Net Operating Cost	94,743	127,816	82,593
Depreciation (20 years Plant Life, Straight-Line)	32,565	38,845	24,440
Return on Rate Base	36,556	43,685	27,626
Federal Income Tax	12,052	14,402	9,107
20-Year Average Annual Revenue Required [†]	175,916	224,748	143,766
Annual Gas Production, 10 ⁹ Btu	79,333	79,596	78,446
20-Year Average Gas Price, \$/10 ⁶ Btu [†]	2.22	2.82	1.83

* This is a nominal coal cost and is not to be interpreted as an IGT recommendation. Depending on mine ownership and capital charges, prices could be in the 40 to 50 ¢/10⁶ Btu range. To avoid establishing a coal cost, its effect has been shown as a variable in Figure 5.

[†] Calculated by the Utility Financing Method (Table 11).

Table 11. GAS COST CALCULATION BY UTILITY METHOD USED IN THE "FINAL
REPORT OF THE FPC SUPPLY-TECHNICAL ADVISORY TASK FORCE - SYNTHETIC GAS-COAL"

<u>BASIS</u>	<u>UTILITY METHOD</u>
Project Life	20 years
Depreciation	5%/year, straight line
Debt/Equity Ratio	75%/25%
Return on Equity	15%
Interest Rate on Debt	9%
Federal Income Tax	48%
Interest During Construction	Interest Rate (9%) X 1.875 years* X total plant investment
<u>OTHER FACTORS</u>	
Plant Stream Factor	90%
Contingencies	15% of installed plant cost
Contractor's Overhead and Profits	15% of total bare cost
Start-up Cost	5% of total plant investment
Working Capital	a) Coal inventory (60 days feed at full rate) b) Material and supplies (0.9% of total plant investment) c) Net receivables at 1/24 X annual revenue required

Derived equation

$$20\text{-year average gas price, } \$/10^6 \text{ Btu} = \frac{N + 0.1198 C + 0.0198 W}{G}$$

Where

N = Net annual operating cost

C = Total capital required

W = Working capital

G = Annual gas production

* 10% for 3 years, 90% for 1.75 years

High-Btu Gas Using HYGAS and U-GAS

The capital required for the HYGAS and the U-GAS plants for producing high-Btu gas are \$674 million and \$805 million (Table 9). Major items in both plants are gasification reactors, purification, oxygen supply, and offsites. The HYGAS reactor system costs more than the U-GAS reactor system because of its greater size, complexity, and the much higher operating pressure required. However, because of the much higher costs for oxygen supply, methanation, synthesis and product gas compression, and steam generation for the simpler U-GAS reactor, total capital investment for the U-GAS Process is \$131 million more than for the HYGAS Process.

The calculated 20-year average gas price of \$2.82/10⁶ Btu when a U-GAS reactor is used for SNG is substantially higher than the price of \$2.22/10⁶ Btu for the HYGAS Process for \$0.50/10⁶ Btu coal (Table 10). A private investor financing method (DCF) was also developed by the FPC task force comprising 100% equity capital, 25-year project life, 16-year sum-of-the-year's digits depreciation, and 12% DCF rate of return. With this method, the gas prices are \$3.63 and \$2.89/10⁶ Btu for the U-GAS and the HYGAS Processes. Use of the U-GAS reactor gives a higher price because of lower conversion efficiency and higher plant cost. This plant requires 11.7 X 10⁶ more coal and produces 10.8 X 10⁶ fewer by-products compared with the HYGAS plant. The by-products of 65.3 long tons/day sulfur, 69.3 tons/day ammonia, and 84,144 gal/day light oil (B-T-X) reduce the HYGAS gas price by about \$0.14/10⁶ Btu at the unit values of \$10/long ton sulfur, \$50/ton ammonia, and \$0.35/gal for the light oil. There are 81.4 long tons/day of sulfur by-product for U-GAS with negligible effect on gas price.

Low-Btu Gas by the U-GAS Process and Its Comparison to High-Btu Gas by the HYGAS Process

Table 9 also shows a total capital investment of \$674 million for the HYGAS high-Btu plant and \$508 million for the U-GAS low-Btu plant. The U-GAS oxygen supply costs \$90 million, twice that for HYGAS. However, all other aspects for low-Btu gas — coal feeding, gasification, product upgrading, and offsites — cost much less.

Table 10 presents annual operating costs, 20-year average annual revenue required, and gas price. HYGAS coal costs are \$8 million/year more than for low-Btu U-GAS; catalyst and chemical costs are \$2.3 million/year more for HYGAS. The U-GAS system requires 25 men/shift fewer in operating labor than the HYGAS system. Capital-related costs are about \$8 million more for the HYGAS system. The higher HYGAS costs are somewhat offset by the \$11 million higher by-product credit. The total net difference in net operation costs is \$12 million. The higher HYGAS capital and operating costs lead to a \$0.39/10⁶ Btu higher gas price for HYGAS (HYGAS \$2.22, U-GAS \$1.83/10⁶ Btu).

If gas price is calculated using the DCF method described above, the U-GAS price is \$2.34/10⁶ Btu compared with high-Btu gas at \$2.89/10⁶ Btu.

Comparison of High- and Low-Btu Gas Price Sensitivities

Figure 4 shows the effect of variations in plant cost on the 20-year average gas price. The effect of variations in both installed equipment cost and total capital cost are shown. An increase of about 67% is added to the installed equipment cost by the various factors used to arrive at total capital required. For a change of \$1 million in installed equipment cost, the gas price varies by 0.36c/10⁶ Btu; for a similar change in total capital required, the gas price changes by

0.22¢/10⁶ Btu, when the utility financing method is used. For the private investor financing method, the numbers are 0.53¢ and 0.31¢/10⁶ Btu. These sensitivity factors apply to all three processes.

Figure 5 shows the effect of varying coal costs on the gas price. For high-Btu gas, the sensitivity is 1.5¢ change in gas price per 1¢ change in coal cost for the HYGAS Process. Because of the lower efficiency, the sensitivity for the U-GAS to SNG process is 1.8¢ change in gas price per 1¢ change in coal cost. The sensitivity for the U-GAS to low-Btu gas process is 1.2¢ change in gas price per 1¢ change in coal cost.

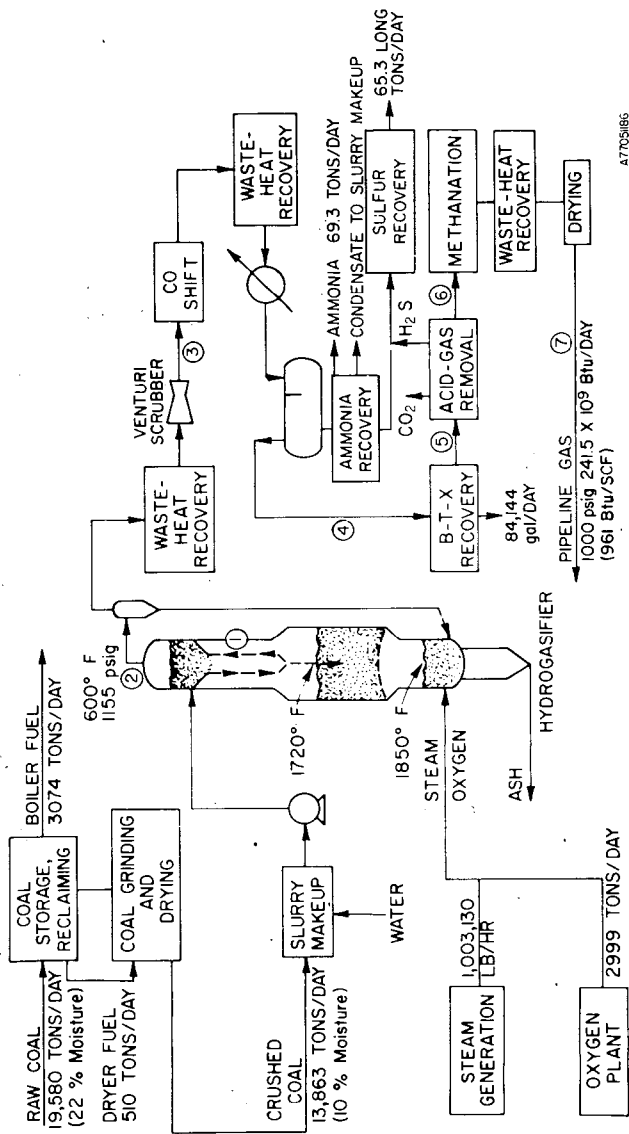
CONCLUSIONS

The manufacture of pipeline-quality gas by the HYGAS Process shows a definite advantage over its manufacture by a single-stage, lower pressure system. Although the hydrogasifier is more complex and operates at a much higher pressure than the U-GAS reactor (1165 vs. 335 psig), a much greater amount of methane is made in the HYGAS reactor. This gives large savings in coal, oxygen, and upgrading costs, resulting in a lower gas price and higher efficiency.

When a low-Btu fuel gas of low methane content is satisfactory, the simpler, low-pressure U-GAS Process shows economic and efficiency advantages.

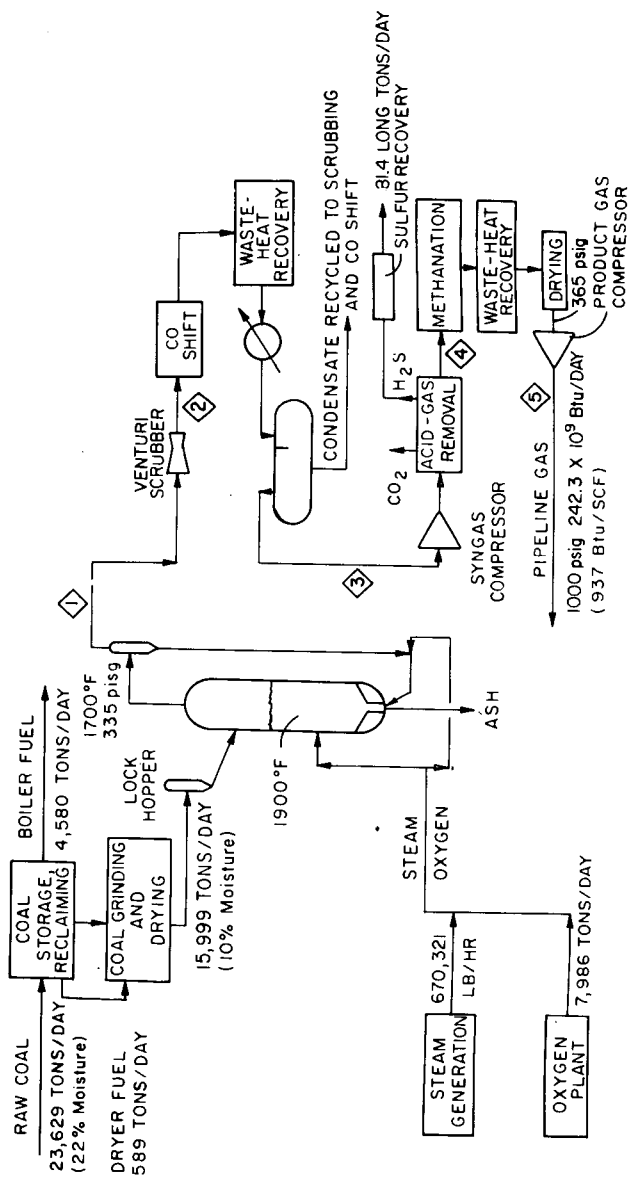
The results are summarized below:

	High-Btu Gas		Low-Btu Gas
	HYGAS	U-GAS	U-GAS
Total capital required, \$10 ⁶ (mid-1976)	674.0	805.0	508.0
Gas price, \$/10 ⁶ Btu, utility financing	2.22	2.82	1.83
Overall thermal efficiency, %	74.0	58.2	80.8



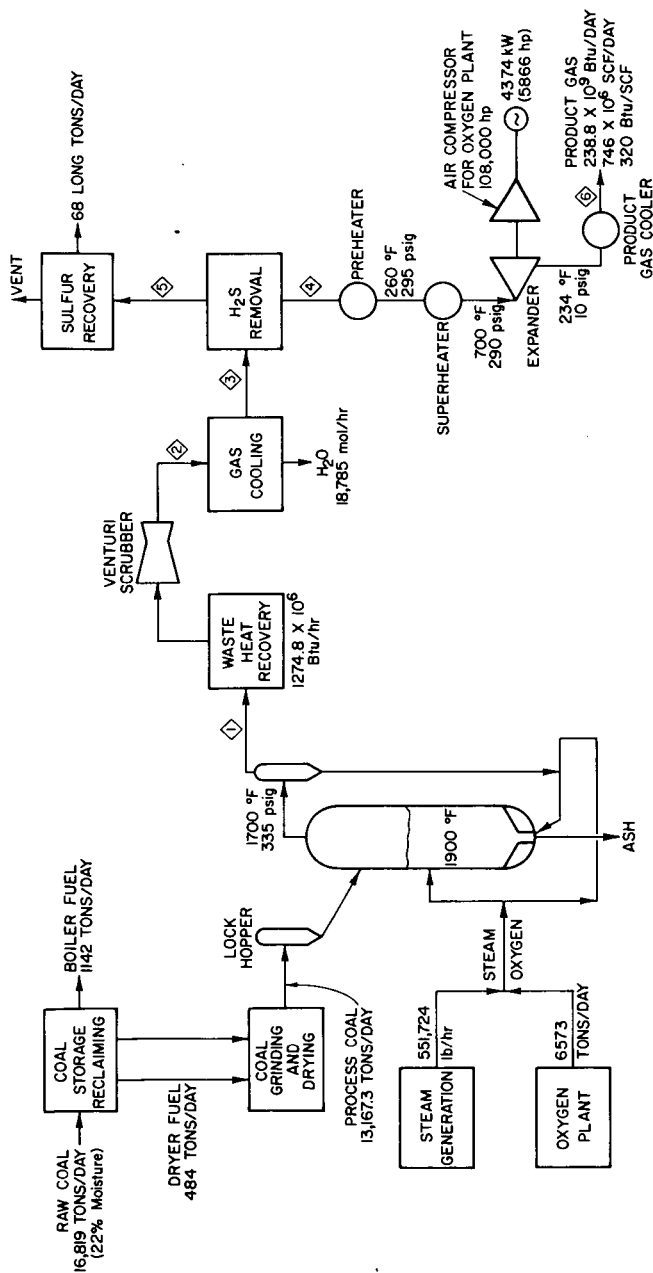
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Figure 1. NOMINAL 240 X 10⁹ Btu/DAY HIGH-Btu GAS BY THE HYGAS STEAM-OXYGEN PROCESS FROM MONTANA SUBBITUMINOUS COAL



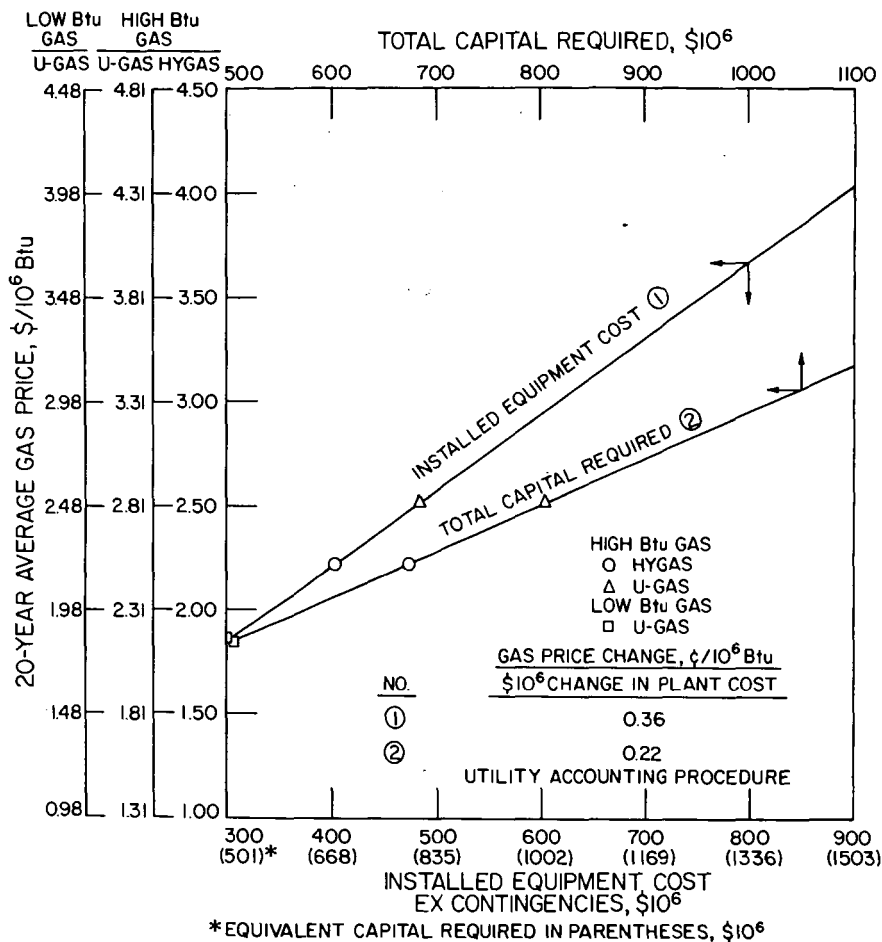
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Figure 2. NOMINAL 240×10^9 Btu/DAY HIGH-Btu GAS BY THE U-GAS PROCESS (OXYGEN-BLOWN) FROM MONTANA SUBBITUMINOUS COAL



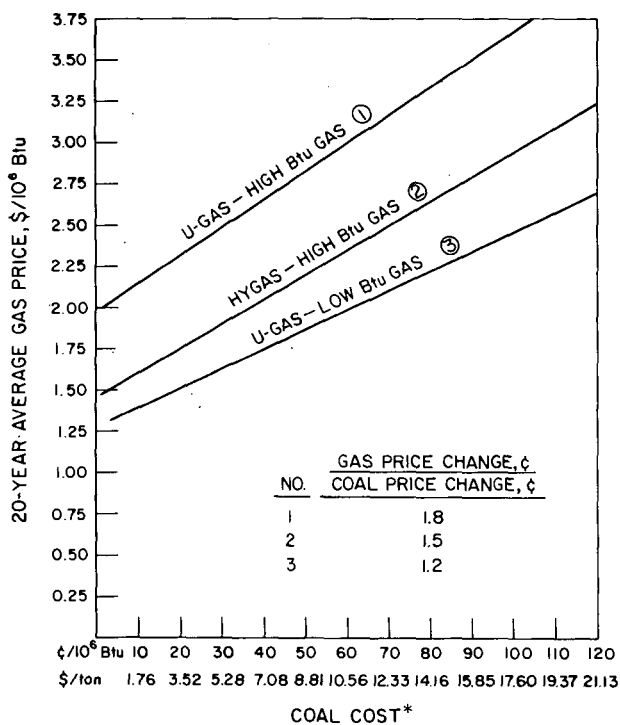
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Figure 3. NOMINAL 240 X 10⁹ Btu/DAY LOW-Btu GAS BY THE U-GAS PROCESS (OXYGEN-BLOWN) FROM MONTANA SUBBITUMINOUS COAL



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Figure 4. EFFECT OF PLANT COST ON GAS PRICE FOR HIGH- AND LOW-Btu GAS FROM MONTANA SUBBITUMINOUS COAL



*11,290 Btu/lb DRY H.V., 22% MOISTURE AS RECEIVED.

A77061257

Figure 5. EFFECT OF COAL COST ON GAS PRICE FOR HIGH- AND LOW-Btu GAS FROM MONTANA SUBBITUMINOUS COAL

COMPARATIVE ECONOMICS OF THE CITIES SERVICE CS-SRT PROCESS WITH THE LURGI PROCESS.
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Cities Service Research and Development Company, the research arm of the Cities Service Company, has been developing a process, The CS-SRT Process, for the non-catalytic, vapor phase, hydrogenation of carbonaceous feedstocks. The initial and primary emphasis in our Energy Research Laboratory was to apply this technology to convert coal into pipeline quality gas and attractive by-products yields of light aromatic (BTX) liquids. Details of the engineering development of this process are reported in another paper at this meeting.

A conceptual plant design for producing 250 MM SCFD of pipeline gas and 6300 BPD of aromatic liquids has been generated. The design was based on data obtained from processing a North Dakota lignite in the bench-scale unit. The design was based on in-house studies and on an engineering study performed for Cities Service by the Foster Wheeler Energy Corporation.

A grass-roots coal conversion plant was designed for a North Dakota-Montana plant site. On-site power generation was included, so that, other than coal feed, only raw water and consumptive chemicals must be supplied. D&E capital investment costs were generated based on fourth quarter 1976 costs. A discounted cash flow analysis was performed to determine the cost of services for producing pipeline gas. The results of the study were compared to a conceptual plant design for producing pipeline quality gas by the Lurgi Process.

ECONOMICS OF PRODUCING METHANOL FROM COAL BY ENTRAINED AND FLUIDIZED-BED GASIFIERS

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Methanol is one of several liquid fuels being considered as a supplement to help alleviate increasing gasoline requirements, the anticipated curtailment of the availability of domestic crude oil and natural gas, and the steadily increasing cost of foreign crude. Methanol has one-half the heating value of gasoline and a lower air requirement for combustion, so some modifications will be required in the carburetor design. Present-day carburetors can operate with a gasoline fuel containing about 10 percent methanol. Coal is being considered as an alternative raw material for the production of methanol owing to the apparent depletion of natural gas supplies.

An economic evaluation of methanol production from coal synthesis gas based on a 5,000-ton-per-day capacity is presented. Two coal gasification systems are considered--entrained and Synthane (a fluid-bed process). Figure 1 is a block diagram showing major units in these two processes. The estimates are based on January 1976 cost indexes. Average selling prices of the gas were determined by using DCF rates of 12, 15, and 20 percent at various coal costs. No inflation factors are included during the life of the plant. Pollution abatement considerations have been incorporated. Some of the economic and technical details are included for the two systems.

ENTRAINED GASIFICATION

In this system methanol is produced from synthesis gas prepared by entrained gasification of Pittsburgh seam coal at 30 atmospheres. (1) Figure 2 is a flow diagram of the process and includes the following steps:

1. Coal preparation, which includes crushing, screening, sizing, and drying of the run-of-mine coal (not shown on figure 2).

2. Entrained oxygen-coal gasification at 30 atmospheres with a 2,400° F outlet gas temperature. Gasifier volume was based on a 2.5-second gas residence time.

3. A dust removal unit removes the entrained dust from the synthesis gas with cyclone separators, before it enters the waste heat recovery unit where steam required in the gasification and shift conversion units is produced. The cooled gas then flows through an electrostatic precipitator for residual dust removal.

4. Shift conversion of the clean synthesis gas to a $H_2:CO$ ratio of 2.3:1 in the presence of sulfur-resistant catalyst. The 50-psig saturated steam required in the purification unit is produced in the heat recovery system following the shift converters.

5. The hot-carbonate purification unit, which reduces the CO_2 content to 2.8 percent and removes essentially all of the H_2S and COS . (2) Char towers are provided for removal of residual sulfur compounds.

6. The low-pressure methanol synthesis system, operating at 570° F and 1,470 psia and utilizing a copper-based catalyst.

It is assumed that approximately 15 percent of the total H_2 and CO entering the converter is synthesized to methanol. The design of the converters was based on a gas space velocity of 10,000 v/v/h. (3) The thermal efficiency of the plant is 46.5 percent, based on a gross heating value of coal at 13,400 Btu per pound and methanol at 10,259 Btu per pound.

SYNTHANE GASIFICATION

The synthesis gas is produced by fluidized gasification of Pittsburgh seam coal at 68 atmospheres. (4) Figure 3 is a flow diagram of the process and includes the following steps:

1. Coal preparation, which includes crushing, screening, sizing, and drying of the run-of-mine coal (not shown on figure 3).
2. Coal pretreatment in the top section of gasification unit to destroy caking properties.
3. Free-fall carbonization plus steam-oxygen gasification of the pretreated coal in a fluidized bed.
4. Entrained char removal in cyclone separators and tar removal by water scrubbing.
5. The first hot-carbonate purification unit, which reduces the CO_2 content to 1 percent and removes essentially all the H_2S and COS . Char towers are provided for removal of residual sulfur compounds.
6. A steam-methane reformer unit, which converts about 95 percent of the CH_4 to CO and H_2 to decrease the amount of purge gas from the methanol synthesis unit in addition to producing H_2 for synthesis.
7. A reverse shift conversion unit, which reduces the $\text{H}_2:\text{CO}$ ratio of the reformer gas product from 4.3:1 to 2.3:1. CO_2 requirement for the reaction is supplied by off gas from the purification system.
8. A second hot carbonate purification unit which selectively reduces the CO_2 content to 2.8 percent. Off gas from the regenerator, namely CO_2 saturated with water vapor, is cooled and compressed to meet CO_2 requirements of the reverse shift converters.
9. The methanol synthesis unit which operates at the same conditions as the other case.

The gasifiers are designed to operate at a pressure of 1,000 psia and at a maximum temperature of 1,800° F with coal throughput of 580 pounds per hour per square foot of cross-sectional area. The assumptions for the methanol synthesis are the same as those used in the entrained gasification system. The thermal efficiency of the plant is 41.6 percent, based on a gross heating value of 13,400 Btu per pound of coal and gross heating values of methanol and byproduct tar of 10,259 Btu per pound and 130,000 Btu per gallon, respectively.

CAPITAL INVESTMENT

The total investment is estimated to be \$331.7 million for the entrained gasification system, or \$252.7 million lower than for the Synthane gasification system.

Table 1 is a capital requirement comparison of the two systems, and figure 4 shows the distribution of capital requirement for major processes. Detailed cost summaries of the major processing units are not included, but the costs of the individual units are listed. General facilities include administrative buildings, shops, warehouses, railroad spurs, rolling stock, roads, waste water treatment, and fences. The cost of steam and power distribution, cooling water towers, plant and instrument air, fire protection, and sanitary water are included in plant utilities.

OPERATING COST

Table 2 presents the estimated operating cost comparison for the entrained and Synthane gasification systems. An assumed 90-percent operating factor allows 35 days for downtime, two 10-day shutdowns for equipment inspection and maintenance, and 15 days for unscheduled operational interruptions. With labor at \$6 per hour, payroll overhead at 30 percent of payroll, and depreciation at 5 percent of the subtotal for depreciation allowing credit for sulfur recovered at \$25 per ton, and with the cost of coal as a variable, the following operating costs are derived:

Cost of coal per ton	Per year, MM		Per gallon		Per MMBtu	
	Entrained	Synthane	Entrained	Synthane	Entrained	Synthane
\$11	\$77.7	\$116.6	\$0.15	\$0.23	\$2.25	\$3.44
13	82.8	122.7	.16	.35	2.38	3.63
15	87.8	128.9	.17	.26	2.50	3.81

Based on a 330-day operation year for the plant and allowing credit for the sulfur produced, with coal costs and discounted cash flow rates as parameters, the average selling prices of the methanol product per gallon and per MMBtu for the two systems are shown in the following table: (These are also plotted on figure 5.)

Cost of coal per ton	Methanol selling price					
	12-pct DCF		15-pct DCF		20-pct DCF	
	Entrained	Synthane	Entrained	Synthane	Entrained	Synthane
	Dollars per gallon					
\$11	0.26	0.43	0.30	0.49	0.37	0.62
13	.27	.44	.31	.51	.38	.63
15	.28	.45	.32	.52	.39	.64
	Dollars per MMBtu					
11	3.84	6.35	4.43	7.23	5.46	9.15
13	3.98	6.49	4.57	7.46	5.61	9.30
15	4.13	6.64	4.72	7.67	5.76	9.44

The DCF computer program takes into account the capital expenditure prior to startup so that the interest during construction is deleted from the capital requirement. Provisions are made for recovery of the working capital in the 20th year.

UNIT COST SUMMARY

The selling price used to determine the high-cost elements in the process was based on a 15-percent DCF for a 20-year project life, with coal at \$13 per ton. A breakdown of the cost elements for the two systems is shown in table 3. Figure 6 shows the selling price of major processes for the two systems.

SUMMARY OF COMPARISONS

As shown in table 1, the total investment for the entrained gasification system is \$252.7 million, or 43 percent lower than for the Synthane system. About 60 percent of the difference is in synthesis gas preparation, consisting of the fluidized gasification system and the methane reforming unit for converting the methane in the product to synthesis gas. It is apparent that the reason for this high capital investment for the Synthane gasification unit is its higher operating pressure; also it requires more separation of such impurities as ammonia, char, and tar from the gas stream. The major part of this capital cost difference comes from the steam-methane reforming unit to process approximately 35 percent (dry base) of methane in the gas stream. This high-temperature reforming reaction is endothermic and requires not only high capital cost but also high-temperature steam to supply the heat required for the reaction. In addition to these, the Synthane system requires extra processes over the entrained gasification system such as a second purification step and CO₂ compression. The higher capital cost of the methanol synthesis unit for the Synthane system, even though both systems have similar feed gas composition and flow rates, is due to the higher compression ratio of the feed gas, resulting in a higher compression cost. Differences in capital costs for these two systems are shown in figure 4.

The operating cost for the entrained gasification system is about 33 percent less than for the Synthane system as shown in table 2. Increases in maintenance, overhead, and indirect and fixed costs, which are directly related to the capital investment, represent the main difference.

The average selling price was based on three coal costs (\$11, \$13, and \$15) and three DCF rates of return (12, 15, and 20 percent). Over this range, the selling price for the entrained gasification is 17 to 25 cents per gallon of product lower than for the Synthane gasification, or \$2.51 to \$3.68 per MMBtu. This represents about a 40-percent decrease.

CONCLUSION

Results of this study clearly indicate the entrained gasification system is more economical than the Synthane fluidized system to produce methanol from coal. The commercial available Lurgi fixed-bed gasification unit will yield similar results owing to the formation of methane in the product as in the fluidized system. One of two alternatives would be a combination of a high-Btu gas plant with methanol as its byproduct. This would eliminate the high cost of a methane-steam reforming unit, but it would have a lower methanol conversion yield because the partial pressure of the hydrogen and carbon monoxide is reduced by the higher methane content in the gas stream plus the fact that the methanol is synthesized on a "once through" basis without recirculation. The other alternative would be the separation of methane from the gas stream by a cryogenic method, but this is also considered a high-capital process. Even though the Synthane coal gasification system may not be the most suitable process to produce methanol from coal owing to its high methane content, it is comparable to other gasification systems that produce high-Btu gas.

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- 2) Wellman, P., and S. Katell. U.S. Bureau of Mines Information Circular 8366, "Hot Carbonate Purification Computer Program," March 1968.
- 3) Danner, G. A. Methanol Technology and Economics. Chem. Eng. Prog. Symposium Series, No. 98, v. 66, 1970.
- 4) Intra-Bureau Report No. 76-17, "Methanol via Synthane Gasification of Pittsburgh Seam Coal, 5,000-Ton-Per-Day Plant." Process Evaluation Office, U.S. Bureau of Mines, Morgantown, W. Va., September 1976.

TABLE 1. - Capital requirements: Comparison of entrained gasification system with Synthane gasification system

Unit	Entrained gasification	Synthane gasification	Difference
Coal preparation.....	\$9,506,900	\$11,327,500	\$-1,820,600
Gasification.....	8,161,200	45,833,000	-37,671,800
Dust removal.....	2,278,900	9,117,000	-6,838,100
Purification No. 1.....	24,731,200	39,728,400	-14,997,200
Methane reforming.....	-	32,462,900	-32,462,900
Waste heat recovery No. 1.	2,169,100	3,599,000	-1,429,900
CO ₂ compression.....	-	23,304,500	-23,304,500
Shift conversion.....	1,629,200	-	+1,629,200
Reverse shift conversion..	-	4,513,900	-4,513,900
Waste heat recovery No. 2.	7,852,200	10,488,100	-2,635,900
Purification No. 2.....	-	29,244,300	-29,244,300
Methanol synthesis.....	63,096,600	86,159,500	-23,062,900
Oxygen plant.....	47,200,000	22,000,000	+25,200,000
Sulfur recovery plant.....	1,100,000	1,130,000	-30,000
Waste water treatment.....	-	10,355,400	-10,355,400
Flue gas processing.....	9,491,400	12,723,700	-3,232,300
Steam and power plant.....	42,452,900	45,595,000	-3,142,100
Plant facilities.....	16,475,200	29,068,100	-12,592,900
Plant utilities.....	23,614,500	41,665,000	-18,050,500
Total construction...	259,759,300	458,315,300	-198,556,000
Initial catalyst requirements.....	2,504,700	3,672,100	-1,167,400
Total plant cost (insurance and tax bases).....	262,264,000	461,987,400	-199,723,400
Interest during construction.....	39,339,600	69,298,100	-29,958,500
Subtotal for depreciation.....	301,603,600	531,285,500	-229,681,900
Working capital.....	30,160,400	53,128,600	-22,968,200
Total investment.....	331,764,000	584,414,100	-252,650,100

TABLE 2. - Annual operating cost: Comparison of
entrained gasification with Synthane gasification

Cost item	Entrained gasification	Synthane gasification	Difference
Direct cost:			
Raw materials:			
Coal at \$11 per ton...	\$27,965,500	\$33,776,400	\$-5,810,900
Raw water.....	691,400	1,375,700	-684,300
Catalyst and chemicals	1,300,200	3,109,700	-1,809,500
Methane.....	143,200	233,400	-90,200
Subtotal.....	30,100,300	38,495,200	-8,394,900
Direct labor.....	2,312,600	2,733,100	-420,500
Direct labor supervision	346,900	410,000	-63,100
Subtotal.....	2,659,500	3,143,100	-483,600
Maintenance labor.....	4,845,000	8,550,000	-3,705,000
Maintenance labor supervision.....	969,000	1,710,000	-741,000
Maintenance material and contracts.....	7,267,500	12,825,000	-5,557,500
Subtotal.....	13,081,500	23,085,000	-10,003,500
Payroll overhead.....	2,542,100	4,020,900	-1,478,800
Operating supplies.....	2,616,300	4,617,000	-2,000,700
Total direct cost..	50,999,700	73,361,200	-22,361,500
Indirect cost.....	7,342,900	12,338,000	-4,995,100
Fixed cost:			
Taxes and insurance.....	5,245,300	9,239,800	-3,994,500
Depreciation.....	15,080,200	26,564,300	-11,484,100
Total, before credit.....	78,668,100	121,503,300	-42,835,200
Sulfur credit.....	994,100	995,900	+1,800
Tar credit.....	-	2,184,200	+2,184,200
Ammonia credit.....	-	1,744,400	+1,744,400
Operating cost, after credit....	77,674,000	116,578,800	-38,904,800

TABLE 3. - Unit cost comparison

Process unit	Cost per gallon of product	
	Entrained gasification	Synthane gasification
Coal preparation.....	\$0.014	\$0.016
Gasification.....	.156	.170
Dust removal.....	.003	.015
Purification No. 1.....	.036	.052
Methane reforming.....	-	.035
CO ₂ compression.....	-	.033
Reverse shift conversion.	-	.005
Shift conversion.....	.023	-
Purification No. 2.....	-	.052
Methanol synthesis.....	.070	.103
Sulfur recovery.....	.001	-
Waste water treatment....	-	.009
Flue gas processing.....	.011	.015
Total.....	.314	.505

NOTE:--Coal at \$13 per ton; DCF at 15 pct.

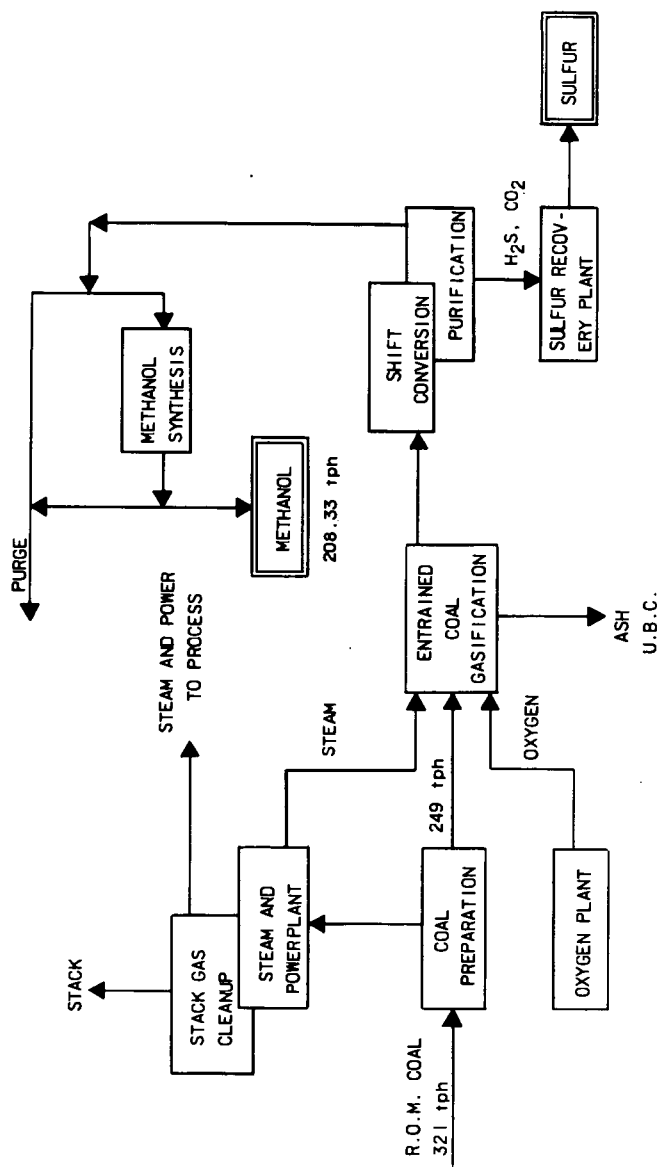


FIGURE 1A. - SCHEMATIC LAYOUT OF ENTRAINED COAL GASIFICATION
TO SYNTHESIZE METHANOL

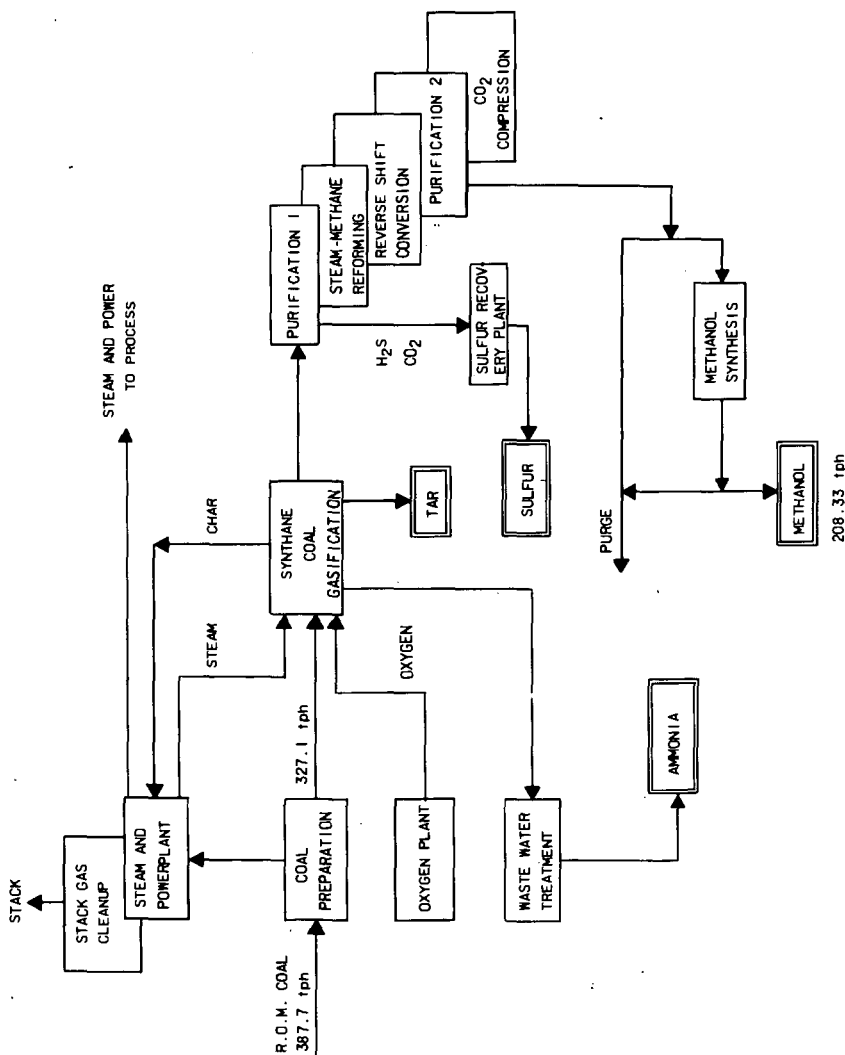


FIGURE 1B - SCHEMATIC LAYOUT OF SYNTHANE COAL GASIFICATION TO SYNTHESIZE METHANOL

208.33 tph

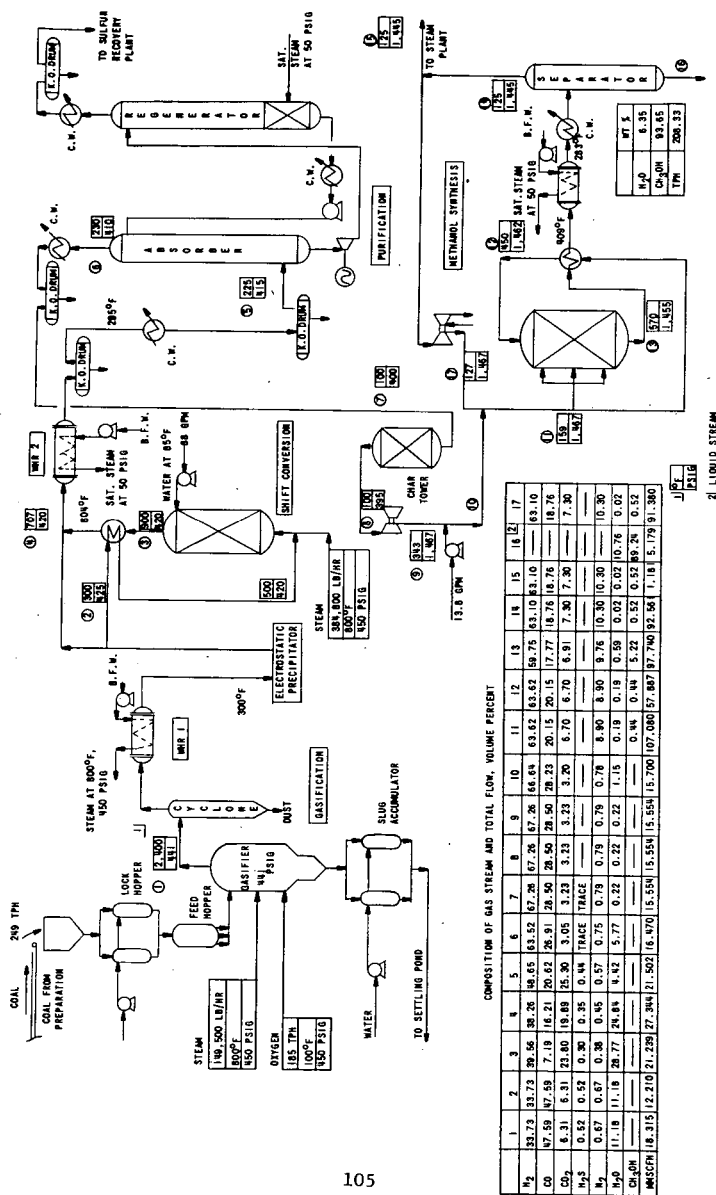


FIG. 2. METHANOL SYNTHESIS VIA ENTRAINED COAL GASIFICATION (5000 TPD)

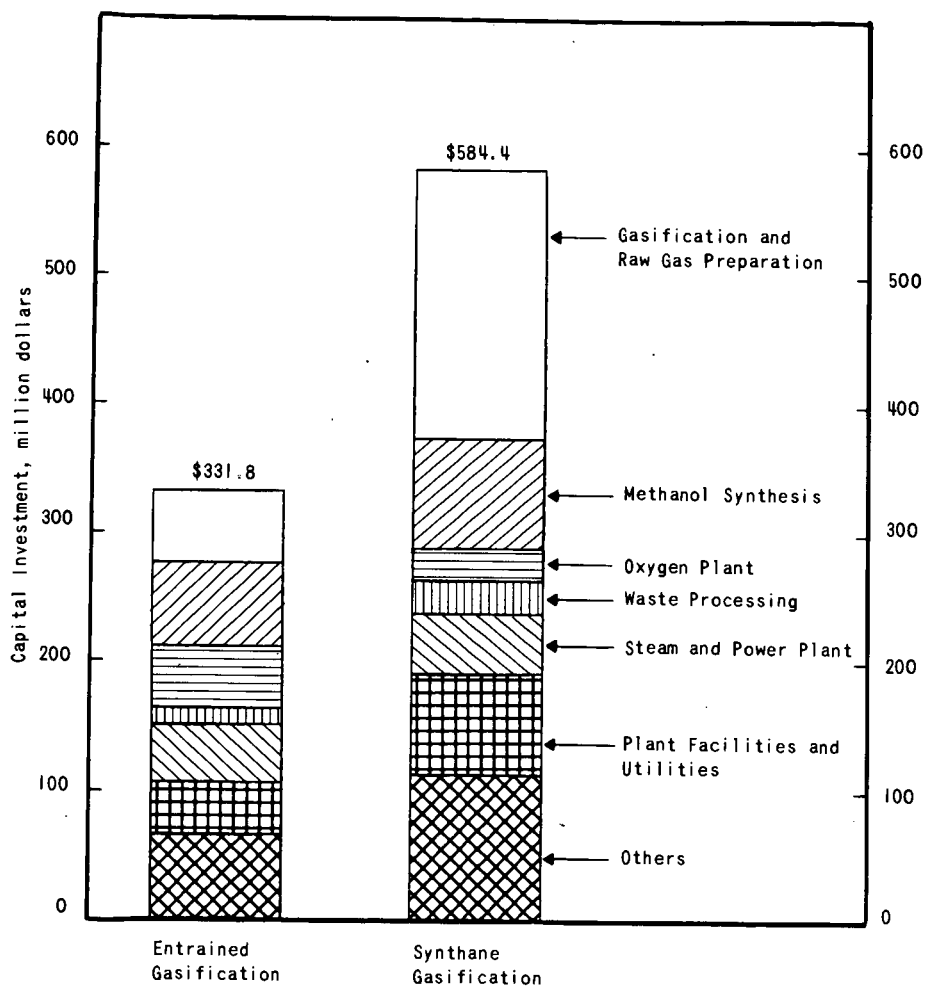


FIGURE 4. - Capital Investment Requirements for Two Processes and Their Distribution

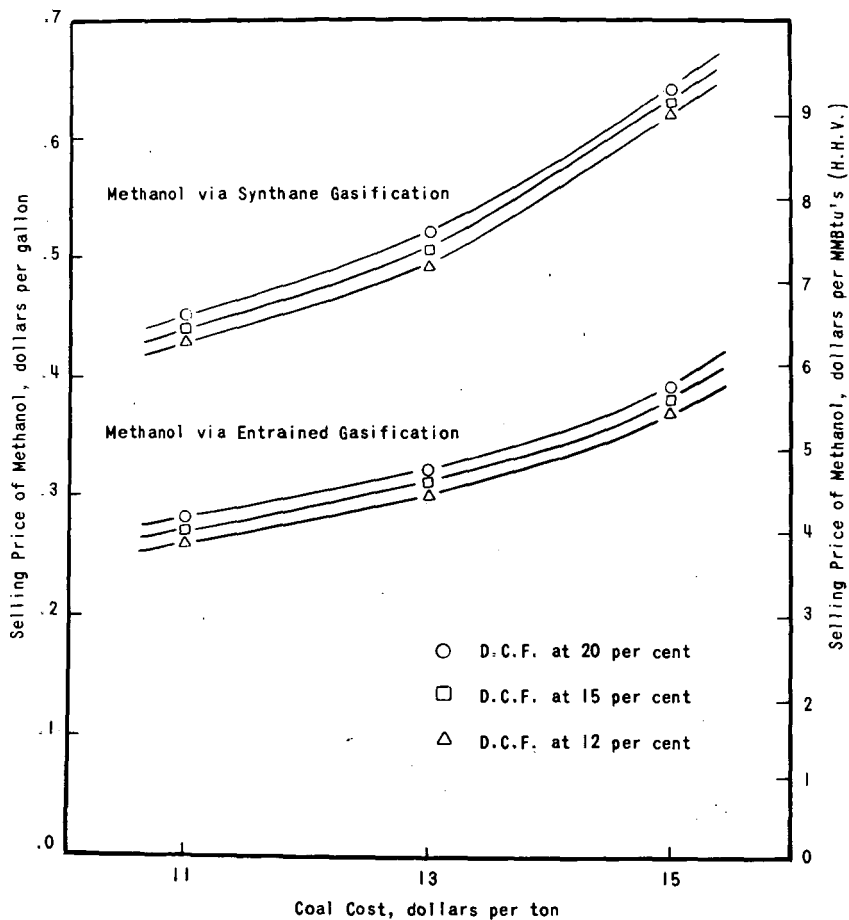


FIGURE 5. - Selling Price of Methanol at Different Coal Prices with D.C.F. Rate of Return as a Parameter

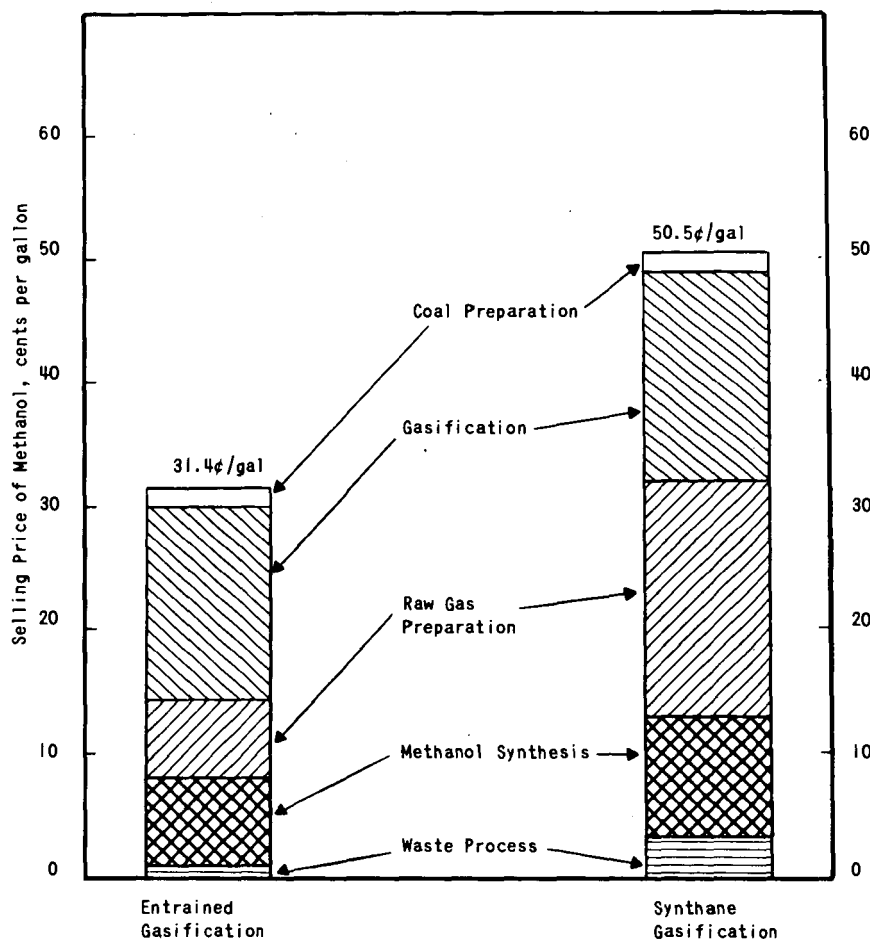


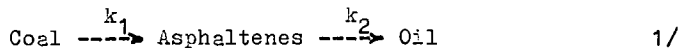
FIGURE 6. - Selling Price of Methanol and its Distribution -
 Calculated Based on Unit Cost Procedure.
 (Selling Prices are Coal at \$13/ton and D.C.F.
 Rate of Return at 15 per cent)

ASPHALTENES AND PREASPHALTENES - COMPONENTS OF AN ORIGINAL hvb COAL

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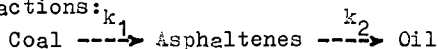
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Asphaltenes are considered to be the principle intermediates in the conversion of coal to an oil products. Weller /1/ stated that the catalytic conversion of coal involves two consecutive first order reactions:



At 400°C k_1 was reported as 27 times higher than k_2 and at 440°C k_1 as 10 times higher than k_2 .

Liebenberg and Potgleter /2/ derived another mechanism which includes the following reactions:



He tentatively determined the sum k_1+k_2 at 400°C and 440°C in the conversion of coal with tetralin and no catalyst added.

More recently Yoshida et al. /3/ established that the mechanism for catalytic conversion of Hokkaido coals includes Reaction 1 and 3. It is worth to emphasize that according to their experiments k_4 is considerably higher or lower than k_1 and k_2 , depending upon coal type.

Sternberg et al. /4,5/ have proposed that preasphaltenes are the intermediates between coal and asphaltenes. Contrary to this, Schwager and Yen /6/ considered that "preasphaltenes may arise from reactive coal depolymerization moieties, which are ... repolymerized into materials more difficult to degrade than the original coal substance".

Collins et al. /7/ claim that "carbon-carbon scission must be considered as an important factor in asphaltene formation" and in preasphaltenes formation as well. The statement has arisen from their experiments based on thermal treatment /over 300°C/ of model compounds - arylalkanes, diphenylalkanes and aryl alkyl ethers.

We have found that asphaltenes and preasphaltenes are the components of coal extracts. Coals were extracted at ambient temperature and precautions in analytical procedure have been observed. Therefore, the components of the extract may be considered they are the components of original coal.

EXPERIMENTAL

High volatile A bituminous coal J /vitritinite 60%, inertinite 33%, exinite 6%/ and hvab coal W /vitritinite 45%, inertinite 45%, exinite 10%/ have been investigated. Proximate and ultimate analyses are presented /Table 1/.

EXTRACTION

Coal samples / <1,4mm/ were extracted in the Soxhlets at ambient temperature for 150 hours. Benzene-ethanol 7:3 vv mixture has been applied. Yields and ultimate analysis of the extracts are given /Table 1/. Although the yields are low they are considerably higher than benzen extract or ethanol extract yields.

Temperature curriculum of the extracted compounds: after extraction at ambient temperature the extract solution has been influenced by temperature below 100°C /the bottom flask of the Soxhlet water bath heated/ for 18 hours /after each 18-hour period, solution has been removed and fresh solvents poured/. The total extract solution was carefully filtrated and the solvents were evaporated from it in a rotary apparatus at 50°C and reduced pressure.

FRACTIONATION OF THE EXTRACTS

has been carried out according to the scheme /Fig. 1/ based on the procedure described by Schweighardt et al./8, 9/ for analysis of hydrogenated coal liquids.

The following group components have been isolated /Table 2/:
preasphaltenes i.e., benzene insolubles/pyridine solubles
asphaltenes i.e., benzene solubles/hexane insolubles
basic fraction of asphaltenes
acidic/neutral fraction of asphaltenes
benzene and hexane solubles

Temperature curriculum of the extracted compounds, continued: all group components were freed from the solvents in a rotary apparatus at 50°C /except pyridine insolubles/ and reduced pressure to constant weight /except pyridine solubles/. Other analytical works were done at ambient temperature. Samples were stored in nitrogen.

Extract+benzene /10g:100ml/ were vigorously stirred for 3 hours. Benzene insolubles BI were filtrated, washed, dried, weighed. Then pyridine was added to BI and total - stirred for 1 hour. Pyridine insolubles PI were filtrated, washed and dried above 100°C to constant weight.

The content of pyridine solubles PS has been determined by difference:

BI - PI = preasphaltenes,
since some final amount of pyridine cannot be removed from them at 50°C.

Solubility of preasphaltenes

We tried to redissolve the preasphaltenes in the same solvent mixture as applied in coal extraction /benzene:ethanol 7:3/. It has been stated that preasphaltenes isolated from the coal extract are easily dissolved at ambient temperature in the mixture, although they are insoluble in benzene or ethanol. Therefore, no indication of their polymerization /6/ which could contribute to a formation of preasphaltenes has been found.

Further analytical work on the separation of preasphaltenes by HCl saturation of benzene/ethanol solution has not been completed yet.

The benzene soluble portion of the extract is reduced in volume by nitrogen flush at 50°C until an approximate ratio 1g:5ml solubles: benzene is reached. Then BS concentrate is slowly introduced by drops into vigorously stirred hexane. Filtrated, washed and dried precipitate yields the content of asphaltenes.

The asphaltenes are dissolved in toluene, filtrated, then dry HCl is bubbled through the solution until no further precipitation is observed. Chloride precipitate is freed from HCl by toluene/1n aqueous NaOH treatment and after evaporation of toluene the content of basic portion of asphaltenes is determined. Compounds which did not form HCl adducts and remained in toluene were recovered by evaporation /nonbasic portion of asphaltenes/.

TLC ANALYSIS OF THE EXTRACT AND ITS SEPARATION PRODUCTS

Table 3. Thin-Layer Chromatography conditions

	Solvent system			Solvent ratio			Develop- ment cycle
	M	C	B	M	C	B	
extract	methanol	chloroform	--	1 : 1	-	-	1/2; full
preasph.	methanol	chloroform	benzene	1 : 1	: 1	-	full
bases	methanol	chloroform	--	1 : 1	-	-	full
nonbases	methanol	chloroform	--	1 : 1	-	-	1/2; full

Neutral gel /type MN-Kieselgel HF/ on 20x20 cm plates was used. Full development cycle was 16 cm high; 1/2 cycle is realized as follows; the plate is kept in the chamber until solvent reached 1/2 height, removed from chamber, dried under nitrogen, reinserted in chamber.

The applied spray reagents for functional group detection on TLC plates are presented /Table 4/.

Table 4. Thin-Layer Spray Reagents

Reagent	Functional group indication				Detection limit / g/
	Ar-OH Phenol type	N-H ring Pyrrol type	=N-ring Pyridine type	Ar-NH ₂ Amines	
Fast Blue Salt B	red- violet	brown- violet	--	yellow- orange	0,01
FeCl ₃ 3% in 0,5n HCl	violet	dark green	--	blue	0,01
Wachmeister's Reagent	yellow- violet	purple- brown	--	brown- green	0,1
Erlich's Reagent	--	violet ^a	light yellow	light yellow	0,05
Dragendorff's Reagent	--	--	orange	pink- red	0,1
Iodoplatinate	--	--	brown	beige	0,5

^a/after few hours

Preparation of the reagents is based on E. Merck handbook - "Anfärbereagenzien für Dünnschicht- und Papier-Chromatographie", Darmstadt, 1970. The same reagents were applied by us /10/ in analysis of hydrogenated coal liquids.

Results of TLC analysis of the coal extract as well as its separation products are summarized /Table 5/.

Table 5. Results of Spray Reagent Test:

	Ar-OH Phenol type	N-H ring Pyrrol type	Functional Group =N- ring Pyridine type	Ar-NH ^a Amine ²
Extract	+	+	+	+
Preasphaltenes	+	+	+	+
Asphaltene bases	---	---	+	+
Asphaltene acidic/neutral	+	+	---	---

a/ TLC-Spray Reagent indication of amine presence is not an irrefutable proof.

b/ Meaningless test - preasphaltenes are contaminated by pyridine during separation procedure.

The results indicate that asphaltenes derived from the coal extract contain heterocompounds which may be separated into basic and acidic/neutral fractions. Phenol and pyrrol derivatives are present in the acidic fraction, pyridine derivatives -- in the basic portion. We did not find any amphoteric substances in the basic nor in the acidic fraction. Therefore, the analogous results have been obtained in analysis of asphaltenes extracted from coal at ambient temperature /this paper/ and asphaltenes derived from the high temperature coal conversion products /5, 8, 9, 10/.

INFRA-RED SPECTROMETRY OF THE EXTRACT AND SOME SEPARATION PRODUCTS

Spectra have been recorded by C. Zeiss spectrophotometer Specord 71 IR Model /NaCl prism/.

Spectra of the extract in KBr pellet /curve A/, basic fraction /curve B/ and acidic/neutral fraction of asphaltenes /curve C/ - both in 2% CS₂ solutions, 0,6 mm fixed path length cell, are presented /Fig. 2/. CS₂ bands have been compensated by application of variable path cell² in reference beam.

Broad and high band in 3590-3050 cm⁻¹ range is a strong indication of the presence of hydrogen bonded heterocompounds in the extract.

Spectrum of bases shows:

in 3570-3100 cm⁻¹ range - weak, broad band which may arise from stretching vibrations of N-H group, free or/and hydrogen bonded; taking into account TLC-spray reagent test results /pyrrol type N-H and phenol type O-H excluded/ the above band arise from amine type N-H bond.

1730 cm^{-1} band indicates carbonyl group of cyclic unsaturated ketones, and/or diketones, and/or quinones. However, this band is absent in the extract spectrum. Therefore, oxidation of the basic fraction may be a reason.

Spectrum of acidic/neutral fraction shows:

in 3585-3550 cm^{-1} range - sharp band characteristic of free phenol O-H and/or intramolecular phenol O-H..... π orbital band;

in 3495-3100 cm^{-1} range - broad band characteristic of inter-molecular O-H bonded group; absorption of pyrrol type N-H stretching and/or NH...OH vibrations occurs within the range as well;

sharp 1030 cm^{-1} band may arise from symmetric stretching vibrations of C-O-C aryl ethers and/or skeletal vibrations of furanes; the presence of furanes is indicated as well by 880 and 860 cm^{-1} dublet.

However, the assignment of all other bands has been carried out, it is not presented here, since complex composition of the samples limits its diagnostic value. On the other hand, part of it is clearly visible and trivial, for instance - higher content of aromatic and alkyl groups in acidic fraction than in basic fraction.

CONCLUSIONS

Asphaltenes and preasphaltenes are the components of an original coal. Therefore, some portions of these group components in coal liquefaction products are not the products of thermal nor catalytic conversion.

The real content of asphaltenes and preasphaltenes in an original coal is still an unanswered question. We have yet to know more on electron-donor and -acceptor properties of solvent mixtures as well as about strength of donor-acceptor bonds occurring in coal substance, if one intends to select solvent mixture capable of extracting total amount of these components from coal.

ACKNOWLEDGEMENT

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TABLE 1. Proximate and ultimate analyses of the coals^{a/}
yields and ultimate analysis of the extracts

	Mad	Ashad	Ydaf	C	H	N	S	O ^{b/}	Yield ^{c/}
Coal J	4,34	4,02	39,05	82,34	5,40	1,64	0,57	10,05	-
Extract from "	-	-	-	82,43	7,45	1,00	0,64	8,48	4,51
Coal W	4,90	9,00	37,40	80,67	5,11	1,40	0,87	11,95	-
Extract from "	-	-	-	82,65	7,08	1,15	1,55	7,57	4,47

a/ Ultimate analysis is given in wt % of daf coal

b/ By difference

c/ Yields of extract are given in wt % of daf coal

TABLE 2. Group composition of coal extracts

Content of the components in /wt %/ :

Group Composition	Extract from Coal J	Coal J ^{a/}	Extract from Coal W	Coal W ^{a/}
Pyridine insolubles /PI/	1,1	0,04	1,0	0,04
Treaspthaltenes /PS/	45,3	2,04	42,7	1,91
Asphaltenes /HI/	23,4	1,06	27,5	1,23
Bases	4,8	0,22	6,7	0,30
Acids + neutral cpds	18,6	0,84	20,8	0,93
Hexane solubles /HS/	22,5	1,02	24,2	1,08
Lost	7,7	0,35	4,6	0,21
TOTAL	100,0	4,51	100,0	4,47

a/ Data are given in weight percent of daf coal

FIG. 4.

EXTRACT FRACTIONATION SCHEME

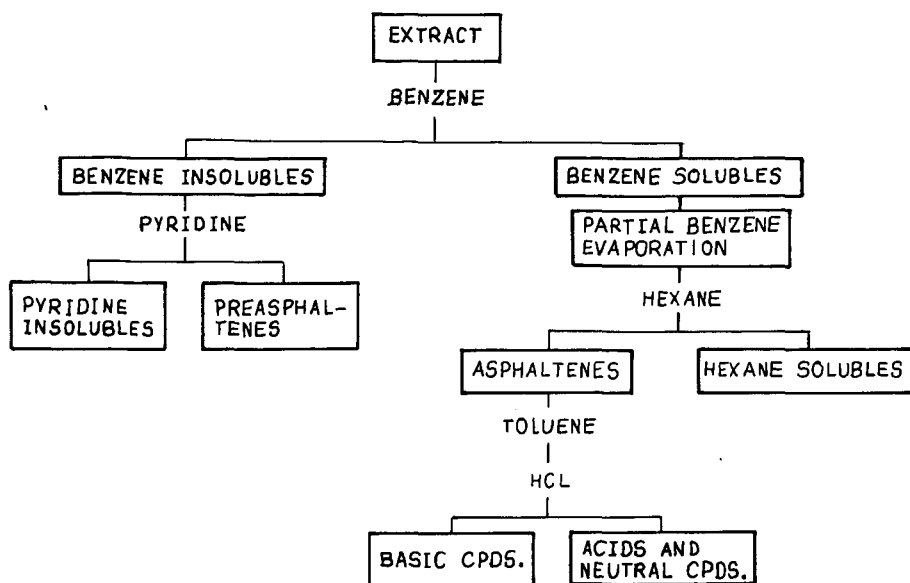
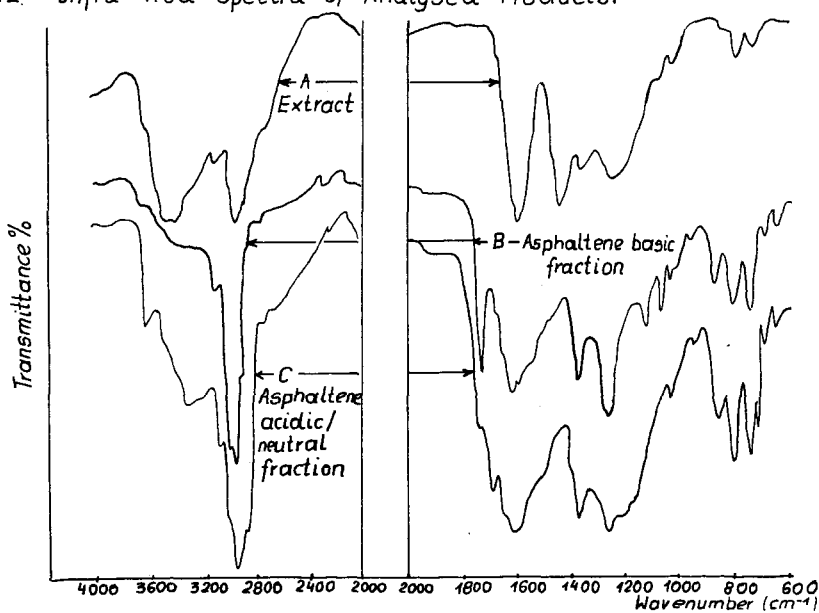


Fig.2 Infra - Red - Spectra of Analysed Products.



THE HYDROTREATMENT OF COAL WITH AlCl_3/HCl AND OTHER STRONG ACID MEDIA

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INTRODUCTION

Most current processes for upgrading coal to cleaner fuels require stringent reaction conditions of high temperatures and pressures. Less severe reaction conditions are needed to make coal upgrading economically feasible. The objective of this work was to investigate catalyst systems for upgrading of coal to clean fuels under moderated conditions. In this work, homogeneous acid catalysts are of particular interest because they allow intimate contact with the coal, they are not liable to coal ash fouling, and they are easily recovered from the coal ash.

The most common homogeneous catalysts studied in coal upgrading belong to the general class of molten salt catalysts, (1-5) and include halide salts of antimony, bismuth, aluminum, and many of the transition metals. Most often, these molten salts have been studied at high temperature, and in massive excess. (1-5) We have performed a systematic study of the use of some of these molten salts as the homogeneous acid catalysts for upgrading of coal at relatively low temperatures and moderate quantities.

In our initial work to establish relatively mild reaction conditions that would still give relatively good conversions, we conducted a series of experiments to determine the role of HCl , AlCl_3 , and H_2 in coal hydrocracking. We examined the effects of temperature and residence time, studied catalyst/coal weight ratios of 1:1 to 3:1, and finally chose the standard reaction conditions for the screening of several acid catalysts. This paper summarizes the experimental studies and our results.

EXPERIMENTAL STUDIES

We used Illinois No. 6 coal, which was pulverized by ball milling under nitrogen to 60 mesh and then usually dried in a vacuum oven at 115°C overnight. Pennsylvania State University supplied beneficiated coal samples (PSOC-26) as well as an unbene-ficiated sample (PSOC-25) for use in some experiments. The reactor used was either a rocking 500-ml autoclave fully lined with Teflon, or a 300-ml Hastelloy C Magne-Drive stirred autoclave from Autoclave Engineers. Standard tetrahydrofuran (THF) and pyridine solubilities were determined for the dried product coal by stirring a 0.50 g sample of the product coal in 50 ml THF or pyridine at room temperature for 1 hr, filtering the mixture in a medium porosity sintered glass filter, and then washing the residue with fresh solvent (~ 50 ml) until the washings were clear.

RESULTS AND DISCUSSION

In a series of runs in a rocking Teflon-lined autoclave, we first studied the role of HCl , AlCl_3 , and H_2 in coal hydrocracking, using 5 g each of AlCl_3 and coal, at 190°C (just above the melting point of AlCl_3), for 15 and 5 hr. As shown in Figure 1, one or more of the three components were absent in Runs 1 to 6 and 9, and in each case no increase in THF and pyridine solubilities was observed. In Run 10, where all three components were present, solubilities increased substantially, suggesting that the AlCl_3/HCl system was active. Runs 7 and 10 serve to assess the importance of HCl in the system under these conditions, however the results are not unequivocal. Here, the presence in the coal of proton sources, such as phenolic groups and traces of water, undoubtedly hydrolyzes some of the AlCl_3 , producing HCl .

These runs indicate that no added HCl is required for coal hydrocracking at these lower temperatures.

At higher reaction temperatures (210°C) and shorter reaction time (5 hr) on the other hand, the added HCl clearly increases the conversion (Runs 21 and 25), suggesting that the effective catalyst in the system must contain the elements of HCl and AlCl_3 .

Next, we studied the effect of potential H-donor hydrocarbons and temperature. We based our work on the results of Siskin, (5) who found that saturated, tertiary hydrocarbons serve as effective hydride donors in the strong acid-promoted hydrogenolysis of benzene. In our system they proved ineffective (Runs 17, 22, 24, and 26, Figure 1). Higher temperatures allowed shorter reaction times. The results for Run 18 (only 5 hr at 195°C) are comparable to those for Runs 7 and 10 (15 hr, 190°C). Runs at 195°C for 15 hr were significantly more effective, and the conversion for Run 12 at 5 hr and 210°C is about the same as that of Run 16 for 15 hr at 195°C.

Next, we studied the effects of the reaction period and catalyst to coal weight ratio on both the product character and the coal product yields in both the Teflon-lined and the Hastelloy C autoclaves. We first compared the effects of the catalyst/coal ratio and at a weight ratio of 1.0, the two systems yielded products with strikingly different pyridine solubilities: about 60% with the Teflon equipment. Increasing the catalyst/coal ratio to 2.0 increased solubilities to above 90%, but a further ratio increase actually caused solubilities to decrease slightly. The solid product recovery also decreased with increasing catalyst/coal ratio. At a 2.0 ratio, only about half the coal was recovered as a solid product. The other half was gasified. The softening point for the THF-soluble fraction was about 150°C; however, the pyridine-soluble fraction did not melt even up to 280°C. The coal products from the Teflon-lined reactor have consistently higher H/C ratio than those from the Hastelloy C reactor. We have no detailed explanation for the effect of autoclave surface on the results, but passivation of the metal surface by some minimum quantity of catalyst is part of the answer. Whatever the mechanism, the Teflon surface is helpful.

In runs over varying residence times in the Hastelloy C autoclave at a constant 2.0 catalyst/coal ratio, we observe significant differences in gasification. In runs of 5 hr down to 90 min, solid product recovery was 45 to 50%. In the 45 min runs, solid product recoveries were 66 to 72%. The H/C values for the isolated solid product coals for all six runs were remarkably similar, from 0.82 to 0.85. Similarly, all coal products have pyridine solubilities greater than 90%. The following scheme may explain these data.

The catalyst system gasifies some of the coal directly to methane and ethane. This result, and the effects of temperature on coal conversion, are shown in Table 1.

The Table shows data from runs at 210°C for reaction times from 45 min to 5 hr, and at 300°C for experiments all for 90 min. (the 210°C data are from an earlier phase of our work, where the gasification was not quantified. Thus the gasification was determined by difference. For the 300°C work, the quantities of gases and residue were determined independently, and thus the mass balances for these runs are not exactly 100%).

From the lower temperature results, it is seen that the degree of gasification increases with increasing reaction time. The solid coal products from these runs are all highly pyridine soluble, and have effectively the same H/C ratio, of 0.83 - 0.84. (The H/C ratio of the starting coal is 0.79).

The 300°C runs are all for 90 min, and experiments 83 and 85 show striking degrees of gasification. Fully 90+% of the carbon in the coal was converted to a

50:50 mixture of methane and ethane in these experiments. The next three runs were run without HCl present, and a cumulative effect of its absence is seen. The degrees of gasification decline severely, and the solid coal products recovered all have lessened pyridine solubilities and H/C ratios. A similar effect for HCl was observed by Kawa, et al.(2).

These data can be explained by the following scheme.

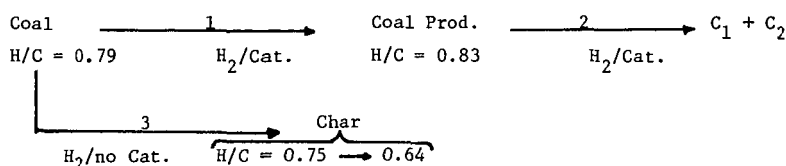


Table 1

THE EFFECT OF RESIDENCE TIMES ON COAL GASIFICATION

(4 g Illinois No. 6 coal, 8 g AlCl_3 , 500 psi HCl, 800 psi H_2 , in a 300 ml stirred Hastelloy C autoclave)

Run	Residence Time	Coal Residue			% Coal Gasified
		% Rcvd	% Pyr Sol	H/C	
210°C					
67	5 hr	39	97	0.83	61 ^b
46 ^c	"	48	87	0.82	52 ^b
41	4 hr	49	91	0.84	51 ^b
66	90 min	47	96	0.85	53 ^b
69 ^d	45 min	72	93	0.82	28 ^b
71	"	66	97	0.84	34 ^b
300°C					
83	90 min	18	78	0.72	96
85	"	18	83	0.74	90
86	"	30	60	0.75	72
87	"	49	31	0.68	56
88	"	68	28	0.64	36

Run Without HCl

^aBased on 4 g of coal.

^bAssumed, based on unaccounted for material.

^cRun with 3 g coal and 6 g AlCl_3 .

Steps 1 and 2 are important in the presence of an effective catalyst and 3 becomes competitive with no effective catalyst present. At lower temperatures, $k_1 > k_2$. Thus, the hydrogen-rich, pyridine-soluble coal product accumulates and can be isolated. At high temperatures, the relative rates of Steps 1 and 2 are reversed, $k_2 > k_1$, and gasification is the major effect. Finally, when the elimination of HCl reduces the catalyst effectiveness, Steps 1 and 2 are suppressed and 3 becomes dominant. Thus

AlCl_3 alone is not only ineffective, it promotes char formation.* Moreover, with the lessening degrees of gasification, the coal residues appear to be increasingly cross-linked and depleted in hydrogen, possibly a result of chemistry at the autoclave surface.

Several acid catalysts were screened in two series of tests at two catalyst concentration levels, 210°C , 5 hr, 800 psi H_2 , and 0.7 M HX (X = Br, Cl, or F). We found catalyst activity to vary considerably from one series to the next (Table 2). Catalyst coal ratio for the first series was 1:1. All catalysts studied at this ratio, except AlBr_3 and AlCl_3 , were ineffective, reducing THF and pyridine solubilities significantly, perhaps because of internal condensation in the starting coal. The coal products in these runs are probably highly cross-linked. AlCl_3 was considerably more effective than AlBr_3 , and HBr alone (Run 30) was, not surprisingly, ineffective. We found HCl to behave similarly (Run 4, Figure 1). Thus, these results indicate a catalyst effectiveness of $\text{AlCl}_3 > \text{AlBr}_3 > \text{SbCl}_3 \approx \text{SbF}_3 \approx \text{ZnCl}_2 \approx \text{TaF}_5 \approx \text{NiSO}_4 \approx \text{CoSO}_4 \approx \text{HBr}$.

Next we ran a more extensive series with the catalysts present at a constant ratio of 0.045 M catalyst/4 g coal, equivalent to 6 g AlCl_3 /4 g coal. Here we found $\text{SbBr}_3 \approx \text{SbCl}_3 > \text{AlBr}_3 > \text{AlCl}_3 > \text{Ni}(\text{AA})_2 > \text{TaF}_5 >> \text{SbF}_5 \approx \text{MoCl}_5 \approx \text{WCl}_6$ (AA = acetylacetonate). In this second series we found first that TaF_5 , which Siskin (6) found to effectively hydrocrack benzene to mixed hexanes, is not at all effective under our conditions. Similarly, ZnCl_2 , the well known coal conversion catalyst, is not effective under these conditions, perhaps because under our relatively mild conditions, ZnCl_2 is not molten (mp, 283°C). Finally, the favorable antimony bromide and chloride results are similar to those reported by Shell (1).

In Run 35 (Table 2), with AlCl_3/HCl , we used unbeneficiated coal. Here, the THF solubility of the product coal increased by almost a factor of 2, to 40%. The pyridine solubility increased slightly, to 66%. Since pyridine is generally a better solvent for coal liquids than is THF, the considerable increase in THF solubility suggests that more lower molecular weight products are obtained when unbeneficiated coal is used. Also, the mineral matter present in the unbeneficiated coal clearly aids in the acid-catalyzed hydrocracking process, suggesting that the mineral matter in the coal is an effective catalyst under acid conditions.

SUMMARY OF RESULTS

We now have established a set of relatively mild experimental conditions for coal conversion and have identified several homogeneous acid catalysts, which under these conditions can convert the coal almost completely to pyridine-soluble material. The reaction conditions for AlCl_3 are 210°C , 45 min, 2:1 AlCl_3 /coal weight ratio, 500 psi H_2 , 800 psi HCl. Also, we have observed that with 1:1 catalyst/coal weight ratio, the effectiveness of the catalysts for conversion is

$\text{AlCl}_3 > \text{AlBr}_3 >> \text{SbCl}_3 \approx \text{SbF}_3 \approx \text{ZnCl}_2 \approx \text{TaF}_5 \approx \text{NiSO}_4 \approx \text{CoSO}_4$.
When a constant molar ratio of catalyst is used, the order is:

$\text{SbBr}_3 \approx \text{SbCl}_3 > \text{AlBr}_3 > \text{AlCl}_3 > \text{Ni}(\text{AA})_2 > \text{TaF}_5 >> \text{SbF}_5 \approx \text{MoCl}_5 \approx \text{WCl}_6$.

We have discovered that adding HCl to AlCl_3 significantly enhances conversion for runs with short residence time (5 hr or shorter) at 210°C . Under similar reaction conditions, a Teflon-lined reactor yields coal products with greater H/C values than does the Hastelloy C reactor. Residence time also has a significant effect on gasification. More than 50% of the coal is gasified for residence times of 95 min or longer; 30% is gasified at 45 min. Finally, we have established that at 300°C reaction temperature, AlCl_3 /coal weight ratio of 2:1, and with HCl added, about 90% of the coal is converted to methane and ethane.

*The cumulative effect of HCl absence is explained by the need for passivated reactor surfaces. When these reactions are run in fully Teflon-lined reactors, the results are significantly improved. Considerably lower catalyst/coal ratios provide results comparable to those shown here.

Table 2
TREATMENT OF ILLINOIS NO. 6 COAL
WITH H₂/STRONG ACID SYSTEMS

Run #	Catalyst System	Pressure (psi)		Solubilities (%) ^b	
		H ₂	HX	THF	Pyr
(a) <u>Constant Weight</u> ^a					
28	AlCl ₃ /HCl/H ₂	800	500	23	58
35	AlCl ₃ /HCl/H ₂ ^c	800	500	40	66
29	AlBr ₃ /HBr/H ₂	980	33g	9	27
27	AlBr ₃ /HBr/H ₂	820	50g	11	32
30	HBr/H ₂	1000	31g	2	6
31	SbCl ₃ /HCl/H ₂	800	500	-	1
32	TaF ₅ /HF/H ₂	1100	22g	-	13
33	SbF ₃ /HF/H ₂	1100	16g	< 1	4
44	ZnCl ₂ /HCl/H ₂	800	500	-	9
56	CoSO ₄ /H ₂ SO ₄	1300	68.6g	< 1	< 1
57	NiSO ₄ /H ₂ SO ₄	1300	68.6g	< 1	< 1
(b) <u>Constant Molar Quantity</u> ^d					
45	AlCl ₃ /HCl/H ₂	800	500	25	47
48	AlBr ₃ /HBr/H ₂	1100	70g	30	59
49	SbCl ₃ /HCl/H ₂	800	500	-	~100
50	TaF ₅ /HF/H ₂	900	14g	11	20
52	SbF ₅ /HF/H ₂	1150	14g	< 1	< 1
54	MoCl ₅	1300	-	8	16
55	WCl ₆	1300	-	6	12
61	SbBr ₃ /HBr	850	59g	43	~100
62	Ni(AA) ₂ /HCl	800	500	ND ^e	38
70	MoCl ₅ /HCl	800	500	< 1	4

^aIn this series of experiments, 5 g of coal was treated at 210°C for 5 hr in a rocking Teflon-lined autoclave. ^b Moisture-ash-free basis. ^c Unbeneficiated coal was used.

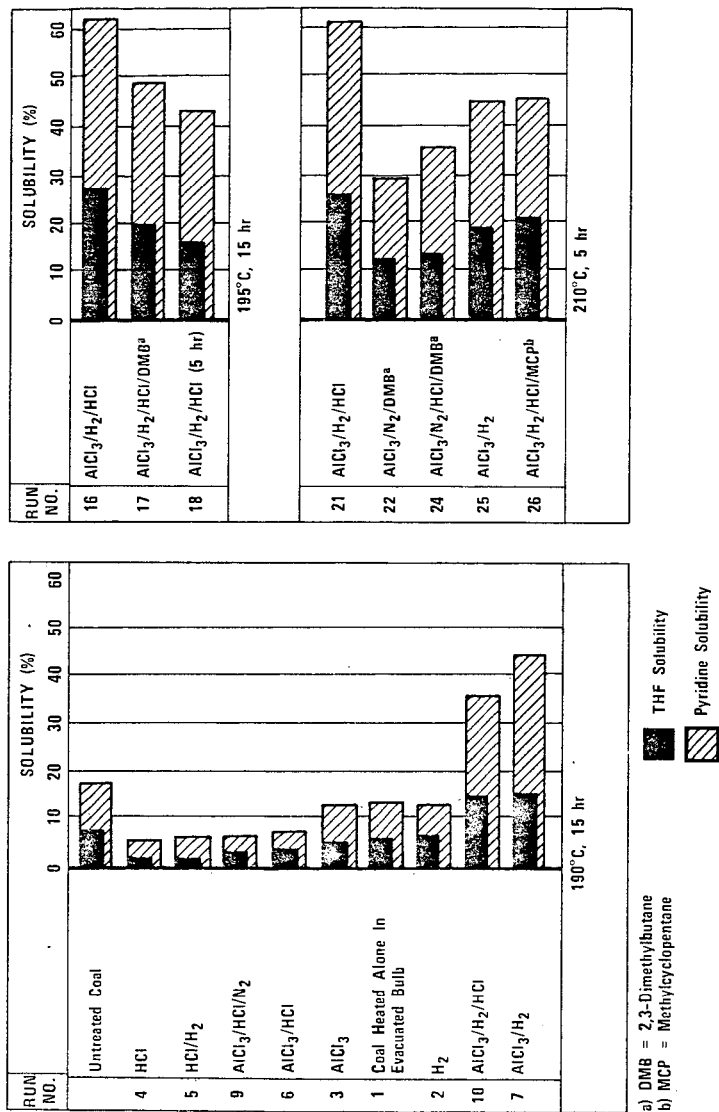
^d These experiments used 0.045 moles of catalyst per 4 g coal. A stirred Hastelloy C autoclave was used. ^e NO = Not determined.

ACKNOWLEDGEMENT

The financial support for this work from Energy Research and Development Administration under Contract No. E(49-18)-2202 is gratefully acknowledged.

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SA-4799-6

FIGURE 1 ACID-CATALYZED HYDROCRACKING OF BENEFICIATED ILLINOIS NO. 9 COAL

Particle Size Analysis in the SRC Process by Coulter Counter

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Introduction

This work was undertaken to develop an effective rapid technique for determining particle size of undissolved solids in the Solvent Refined Coal (SRC) Process. Particle size distributions for SRC filter feed from the Wilsonville SRC Pilot Plant and autoclave reaction mixtures from the Auburn Coal Conversion Laboratory were measured using a Coulter Counter Model TA.

The Coulter principle was originally applied to blood cell counting¹ and is now widely used in the biomedical area for cell counting and size distribution.² Industrial use of the Coulter technique is now widespread.³ The Coulter technique has been applied in coal research in the size analysis of pulverized coal, coal dust, and fly-ash.⁴ Presently, the Coulter technique is being applied to the study of particle size distributions in the SRC process. Interest in the effect of the mean particle size in the effluent from the SRC dissolver on filtration along with the problem of dissolver solids accumulation⁵ has shown the need for a reproducible method of determining particle size in SRC process streams.

Experimental

Equipment

Particle size determinations were performed on a Coulter Counter Model TA (Coulter Electronics, Hialeah, Florida) equipped with a Model TA External Size Calibrator II and an adjacent sample stand. The orifices normally used for the analyses were 100, 140, 200 and 280 μm .

Electron microscopy was performed using an AMR Model 1000 scanning Electron Microscope operating at 20KV with a tungsten source and an aluminum coated detector. The samples were coated with 60:40 gold-palladium using a Denton DV 502 vacuum evaporator. Optical microscopy was performed using a Wild Model M21 microscope equipped with a Polaroid MP-3 Land Camera.

Chemicals

Reagent grade ammonium thiocyanate (NH_4SCN) and dimethylformamide (DMF) obtained from Mallinckrodt prepared in a 5% solution was the electrolyte used for Coulter analysis. Metrical alpha -8 filters with 0.20 μm pore size were used for filtering the electrolyte solution. Carbowax 400 obtained from Union Carbide was the dispersant used for optical microscopy.

Procedure

Calibration of each orifice was performed using polystyrene calibration standards of known diameter (supplied by Coulter Electronics) dispersed in isoton.

Autoclave reaction samples and Wilsonville filter feed were sonicated using a low energy ultrasonic bath, dispersed in the 5% NH_4SCN - DMF electrolyte and again sonicated. The samples were diluted with electrolyte and immediately analyzed. For each sample 100,000 particles were counted and the particle size distribution was obtained. Extensive filtering with 0.2 μm filters was necessary to maintain the necessary background of the electrolyte.

For scanning electron microscopy samples were prepared by sonicating and further diluting the Coulter samples. The particles were filtered on a 0.2 μm filter and air dried. For optical microscopy, autoclave reaction mixtures were first sonicated and then dispersed in Carbowax 400. Hanging drop as well as slide preparations were used.

Verification

Verification of the Coulter technique by optical microscopy can be seen in Figure 1. Rigid polystyrene calibration spheres (9.99 μ m in diameter supplied by Coulter Electronics) were measured by optical microscopy under 500x magnification to have a diameter of 10.0 μ m. Subsequent analysis of the polystyrene's particle size by Coulter Counter agreed fully with the microscopy.

Results and Discussion

The Coulter technique measures the particle volume size distribution. The smallest particles measured had a diameter of 1.59 μ m with an overall diameter range of 1.59 to 128 μ m. All measurements were taken within the optimum range of the orifice. Particle size distributions were obtained for autoclave reaction mixtures and Wilsonville filter feed of Amax, Western Kentucky, Pittsburgh Seam, and Monterey coals. The particle size distribution measurements for each coal sample can be accurately reproduced within $\pm 10\%$. A mean particle size is obtained graphically by plotting the normalized cumulative particle volume percent versus particle diameter and then obtaining the mean particle size at the 50% volume level.

The initial mean particle size for unreacted coal was between 28.8 - 40.0 μ m. Table I shows the mean particle size in terms of particle diameter for four autoclaved coals reacted at 410°C for three different time intervals. Under autoclave conditions, the mean particle size varies significantly among the different coals. Within the time range studied the mean particle size for Monterey and Pittsburgh Seam coals remained essentially constant; however, the mean particle size of Western Kentucky coal increased with time while that of Amax coal decreased. The mean of Amax coal leveled off at 9.6 μ m after 4 hours. The temporal behavior of the particle size of each coal can be directly correlated to its dissolution rate and behavior. Both Monterey and Pittsburgh Seam coals dissolve very rapidly, leaving essentially only mineral matter after 15 minutes of reaction. Amax coal, however, is a slow dissolver which accounts for the decrease in particle size over the time period studied. Western Kentucky coal dissolves very rapidly but is sticky and adhesive; increasing reaction time allows the particles time to adhere to one another forming larger particles.⁶

The Wilsonville filter feed particle size distributions for each coal closely resembled the autoclave reaction distribution (an example is shown in Table II) except for Western Kentucky coal which showed a significant shift to smaller particles. Generally, the Wilsonville filter feed distributions showed greater variation within each sample than did the autoclave reactions resulting in a larger standard deviation for each volume range. A comparison of the mean particle size of Wilsonville filter feed to Auburn autoclave reaction mixtures is shown in Figure III. For each coal the mean particle sizes compare very closely with the exception of Western Kentucky coal. This difference can again be attributed to the adhesive nature of the Western Kentucky particles. In the filter feed the particles will tend to adhere to one another forming larger particles which stay in the dissolver while the smaller particles are elutriated out of the dissolver.

Optical and electron microscopy verified the presence, size, and character of the particles being counted by the Coulter Counter. Optical microscopy allowed observation of the particles in a solution matrix. Scanning electron microscopy permitted isolation of the particles from the oil matrix and allowed the particles to be individually observed. The unregular nature and individual character of the particles are shown in the micrographs in Figure 2-7.

Process Applications

Filterability of a solid/liquid slurry depends on the size distribution of the particulates entrained in the slurry as well as other factors such as viscosity, adhesive forces among the particles (i.e., whether the particles are sticky or discrete and unattractive) etc. A major objective of this work is to

determine whether mean particle size determined by Coulter Counter analysis can be correlated to filterability or, more specifically, cake resistivity. To this end: the mean particle size of particulates in the SRC dissolver effluent for the coals studied are compared with their associated cake resistivities.

Filtration rates for Amax, Western Kentucky and Monterey coal/oil slurries have been extensively studied under test conditions and without the incorporation of bodyfeed at the Wilsonville SRC Pilot Plant. Pittsburgh Seam coal/oil slurries have only been tested with bodyfeed. Table IV shows the cake resistivities calculated for the slurries run without bodyfeed. Indeed, correlations are evident between the ease of filtration (i.e., cake resistivity) and mean particle size. Amax coal/oil slurries which have the lowest cake resistivity and are, therefore, the most easily filtered of the three types of slurries considered have the largest mean particle size. Monterey coal/oil slurries have the highest cake resistivity, are the most difficult to filter, and have the smallest mean particle size. When screen blinding is taken into account, Western Kentucky ranks second in filterability as well as mean particle size. Cake resistivity is a function of the porosity of the cake which is directly related to particle volume. Particle volume is measured directly by the Coulter technique. In conclusion, the Coulter Counter can give a good indication of the filterability of a particular SRC process stream and, thus, should prove to be a useful tool in optimizing operational conditions for solids separation.

The present Coulter technique has two main limitations. First, since particles less than $1.6\mu\text{m}$ are difficult to detect, the total particle size range of the SRC effluents cannot be studied. The effect of the very small particles on filterability is not precisely known; however, it is very likely that particles below $2\mu\text{m}$ play a significant role in screen blinding. The second limitation is that carbonaceous agglomeration of SRC itself is difficult to detect because approximately 96% of SRC dissolves in the electrolyte. Detection of the formation of solid agglomerates would be highly desirable; for the accumulation of solids in the SRC dissolver has presented significant problems in processing certain types of coals, particularly those containing high concentrations of calcium. One reason for this accumulation, however, is commonly thought to be the formation and growth of calcium carbonate (CaCO_3) crystals. Inorganic agglomeration due to CaCO_3 crystal formation should be easily detectable by the Coulter technique presented herein since CaCO_3 is essentially insoluble in DMF.

On-line Coulter analysis of the SRC process stream would allow immediate detection of changes in the size distribution of the solvent stream particles. Through on-line analysis more precise correlations between mean particle size and filterability would be possible. Also, by monitoring the size distribution of particulates in the upstream and downstream to the filter, a direct reading on filtering efficiency could be obtained. Coulter analysis is also an effective method for determining the efficiency of other solid-liquid separation techniques, such as centrifuging and hydrocloning, currently being tested in the SRC process.

Acknowledgements

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TABLE I
Mean Particle Size of Autoclave Reacted Coals
at 15, 30 and 60 Minutes

Autoclave Conditions				
Temperature: 410°C				
Pressure: Hydrogen 2000 psi				
Solvent/Coal: 3/1				
Stirrer: 1000 rpm				
Time (min)	Mean Particle Size (μm)*			
	Amox	Western Kentucky	Monterey	Pittsburgh Seam
15	14.0	7.0	5.2	6.0
30	9.1	8.2	5.5	5.5
60	8.0	10.6*	4.8	6.0

*Through subsequent calculations, the population mean can be calculated. For example, Western Kentucky coal, reacted for 60 minutes, has a population of 2.0 μm .

TABLE II
Particle Size Distributions of Amax Coal Autoclave
Reaction Mixture and Wilsonville Filter Feed

Particle Diameter (μm)	% Volume Distributions	
	Amax Autoclave 60 minute	Amax Wilsonville filter feed
1.59	10.9 \pm 0.6	10.4 \pm 1.8
2.00	8.7 \pm 1.2	6.3 \pm 1.4
2.52	6.6 \pm 0.64	5.7 \pm 0.6
3.17	5.5 \pm 0.35	6.7 \pm 0.7
4.00	5.6 \pm 0.36	7.6 \pm 0.8
5.04	6.4 \pm 0.42	9.3 \pm 0.7
6.35	6.9 \pm 0.49	8.6 \pm 1.0
8.00	8.2 \pm 0.57	8.8 \pm 1.1
10.08	9.0 \pm 0.68	9.2 \pm 1.3
12.7	9.7 \pm 1.6	9.2 \pm 1.7
16.0	9.2 \pm 1.5	5.7 \pm 0.8
20.2	7.0 \pm 1.8	4.6 \pm 2.0
25.4	4.6 \pm 2.6	4.6 \pm 2.8
32.0	1.3 \pm 1.0	2.3 \pm 2.7
40.3	0.6 \pm 0.05	0.6 \pm 0.1

TABLE III
Comparison of the Mean Particle Size
of Wilsonville Filter Feed and
Auburn Autoclave Reaction Mixtures

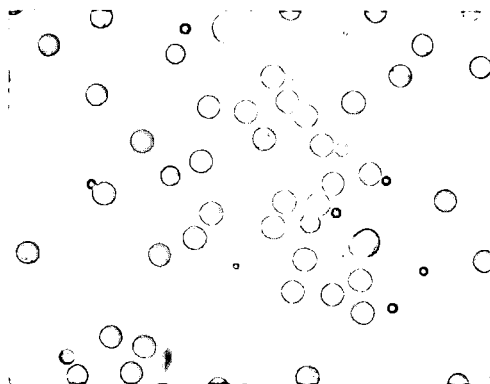
Coal Type	Date	Wilsonville Filter Feed Mean Particle Size	Auburn Autoclave Mean Particle Size
		μm	μm
Monterey	10-20-76	4.5	
Monterey	11-15-76	3.7	
Monterey	11-10-76	3.8	
Monterey	Average	4.0	4.8
Amax	- - - -	7.2	
Amax	12-19-76	6.4	
Amax	1-28-77	7.5	
Amax	Average	7.0	8.0
Western Kentucky	4-17-76	6.8	
Western Kentucky	5-29-76	4.6	
Western Kentucky	Average	5.7	10.6
Pittsburgh Seam	7-22-75	6.2	6.0

TABLE IV
Correlation between Cake Resistivity and Mean
Particle Size of Wilsonville Filter Feed

Coal	Cake Resistivity ft/lbm	ΔP (psi)	Mean Particle Size (μm)	
Amax	0.8×10^{12}	45	7.0	(Ref 7)
Western Kentucky	1.13×10^{12}	60	5.7	(Ref 8)
Monterey	1.22×10^{12}	60	4.0	(Ref 8)

Figure 1

Verification of
polystyrene particle
size



1 cm = 20 μm

Figure 2

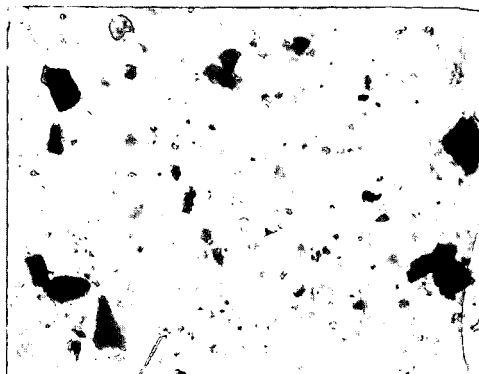
Photomicrograph of
Amax coal particles
(suspended in Carbowax)
after 60 minute autoclave
reaction (500x)



1 cm = 20 μm

Figure 3

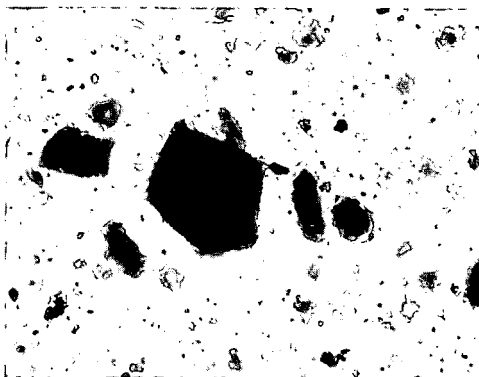
Photomicrograph of
Pittsburgh Seam coal particles
(suspended in carbowax) after
60 minute autoclave reaction
(500x)



1 cm = 20 μ m

Figure 4

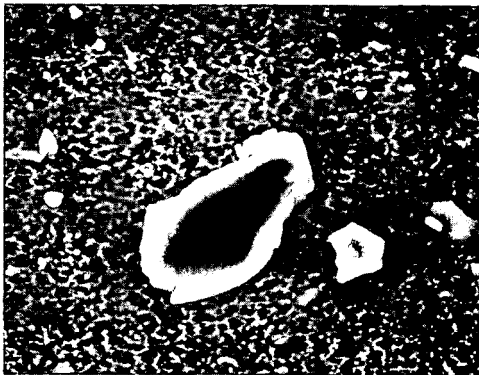
Photomicrograph of
Western Kentucky coal particles
(suspended in carbowax)
after 60 minute autoclave
reaction (500x)



1 cm = 20 μ m

Figure 5

Electronicrograph of
Western Kentucky coal particles
(on a Metrice1 filter)
after 60 minute autoclave
reaction (2000x)



1 cm = 5 μ m

Figure 6

Electronmicrograph of
Monterey coal particles
(on a Metrice1 filter)
after 60 minute autoclave
reaction (2000x)



1 cm = 5 μ m

Figure 7

Electronmicrograph of
Monterey coal particle
(on a Metrice1 filter)
after 60 minute autoclave
reaction (10,000x)



1 cm = 1 μ m

ENGINEERING DEVELOPMENT OF THE CITIES SERVICE,
SHORT RESIDENCE TIME (CS-SRT) PROCESS

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History and Development of the CS-SRT Process

Cities Service Research and Development Company (CSRD), the research arm of the Cities Service Company, has been developing a process scheme named The CS-SRT Process, for the non-catalytic, vapor-phase, hydrogenation of carbonaceous feedstocks. The initial and primary emphasis in our Energy Research Laboratory was to apply this technology to convert coal into pipeline quality gas and attractive byproduct yields of light aromatic (BTX) liquids.

A review of the literature early in 1974 led to our conclusion that the most productive area of process development would be that of short residence time hydrogenation. A brief literature review has been presented in our previous paper at the 1976 ACS (San Francisco) meeting (1).

In the summer of 1974, we designed a 1-4 lb/hr bench-scale unit capable of operation at temperatures and pressures that are scalable within present-day, commercial technology. A cold-flow model to test coal-hydrogen mixing injectors and to study coal-hydrogen slip velocities was designed and constructed in Autumn, 1974. Construction of the bench-scale unit began in December, 1974 and was completed in June, 1975 with the shakedown operations occurring in the Summer of 1975. The first complete material balance run was made in August, 1975. Since that time, over 125 runs have been made with a variety of feedstocks including lignite, bituminous and subbituminous coals, oil shale, tar sands and coal tars.

In September, 1974 work was initiated to estimate the economics in a conceptualized, commercial application of the CS-SRT Process for producing 250 MM SCFD of pipeline gas (and the associated byproduct BTX-liquids) from a mine-mouth plant in the Montana/North Dakota region. The results of this work pointed out the dramatic effect of benzene byproduct yield on lowering the cost of services for producing pipeline gas. In September, 1976, CSRD retained the Foster Wheeler Energy Corporation to perform a preliminary engineering design and cost study of the CS-SRT Process to validate the results of the earlier economic study. The results of this study will be reported in a separate paper.

Further development of the CS-SRT Process is being undertaken in several programs supported jointly by Cities Service, Rocketdyne Division of Rockwell International, and ERDA. Experimental tests are continuing in the bench-scale unit to explore conditions for maximizing liquids yields and also for maximizing gas (methane, ethane) yields. Process flowsheet studies are also being made. The next step of development of the CS-SRT Process would require about a 6-inch diameter, pilot plant reactor capable of processing about 100 TPD coal. This pilot-plant would be one scaleup step away from a 12-inch diameter, single tube, commercial-type operation.

This paper summarizes the analysis of the bench-scale data obtained when processing a North Dakota lignite. Some conceptual commercial processing alternatives which appear quite attractive are also discussed.

Experimental Apparatus

The bench-scale unit, as described in our previous paper, utilized initially a dilute-phase, free-fall reactor with a movable quench probe. Coal was delivered to the hydrogen-coal mixing injectors under gravity flow from a star-wheel, volumetric feeder fed by a lock hopper. The feeder-hopper assembly sat directly on top of the reactor.

Since the initial testing, a coal transport system was developed to improve the running efficiency of the test program. This system also allowed for the remote location and safe access of the coal feeding equipment in an adjacent laboratory pressure cell. The transported coal was separated in a cyclone immediately upstream of the coal-hot hydrogen injectors and carried into the reactor. The "cold" hydrogen was recycled back to a diaphragm compressor. The compressor diaphragms were protected against entrained solids by use of a settling chamber and filters located on the suction-side of the compressor.

Several other reactor designs were also incorporated into the bench-scale unit since the initial testing. A description of these reactors is shown in Figure 1. In these "entrained-flow" reactors, a wider range of values of several important parameters is achieved over that of the free-fall reactor:

	<u>Free Fall</u>	<u>Entrained-Flow</u>
Coal Flux, lb/hr ft ²	100-300	1500-35,000
Gas Velocity, fps	0.1-0.5	2-50
Particle Velocity, fps	0.5-8	2-50
Gas Reynolds No.	100-500	1000-6000
Particle Residence Time, sec.	0.10-4	0.05-10
Vapor Residence Time, sec.	2-30	0.05-5

The vertical entrained-flow reactor is used generally for very short residence time studies (less than 200 milliseconds) whereas the helical entrained-flow reactors are used for longer residence times (c.a., 1-5 sec.). All of the reactors used in the bench-scale unit are fabricated from stainless steel 316 alloy. A "hot wall" pressure reactor was used as the basis of design to expedite the testing program. The life of the hot wall reactor, even at the small diametral ratios used, is limited because of operation at high stress levels. Therefore, careful watch is maintained on the instantaneous creep strength of each reactor and preheater vessel. Reactors are removed from service before the cumulative approach equals 25% of the rupture time. All reactors are operated in the confines of a high-pressure, explosion-proof, laboratory cell with lockout controls during testing.

Temperature and Residence Time Measurements

Temperature of the coal-hydrogen stream is an extremely important variable affecting the rates of hydrolysis under short residence time conditions. Most experimenters involved in continuous, two-phase, coal-gas reactions report reactant or reactor temperature as maximum reactor wall temperature. This latter temperature can vary quite markedly from the reactant temperature depending upon the size and mass of the reactor, coal throughput, exothermicity of reaction, heat losses, etc. Accordingly, we have developed several multi-dimensional, computerized, heat transfer models for estimating reactant temperatures from reactor wall thermocouple readings. We have also used equivalent isothermal temperature (EIT) calculations to characterize the non-isothermal reaction conditions. EIT has been found to be a good characterization factor in correlating trends of hydrolysis yields with temperature.

Particle residence time is another extremely important parameter affecting hydrolysis yields, particularly under short residence time conditions. In the case of the free-fall reactor, particles traverse the reactor under the influence of both gravity and entrainment by hydrogen. Char settling velocities were calculated using the Stokes equation for narrow-size ranges of particles. Corrections were applied for the wider-range size distributions used in the bench-scale operation by calculating a surface-volume average particle diameter. Based on studies by Wen and Huebler (2), corrections for gas entrainment, uneven particle distributions and particle cloud density were also made.

For the case of entrained-flow reactors, particle residence times are directly correlated with the superficial hydrogen velocities. For both cases, a comprehensive experimental (cold-flow) program is underway in order to measure accurately particle residence times in both free-fall and entrained-flow reactors.

Coal Preparation

Wet North Dakota lignite was received in drums from the Grand Forks Energy Research Center. Prior to drying, the lignite was reduced to 1/2-inch x 0 particle size using a rotary jaw mill blanketed with nitrogen. Drying was done either in a laboratory, fixed-bed drier at 15" Hg vacuum or in an industrial, fluid-bed dryer, both under nitrogen blanketing. In both cases, the maximum lignite temperature was limited to 230-250°F. Moisture was reduced from 35% to less than 5%. The lignite was then pulverized using either a Sweco ball mill or a Mikropul hammer mill. In both cases, a nitrogen atmosphere was maintained. An 18-inch, continuous Sweco sieve, fed by a Syntrol vibrating feeder, was used to sieve the dried, pulverized lignite to specification.

Immediately prior to a run, the lignite charge to the unit was dried again, under vacuum, in the laboratory tray drier. An aliquot sample from a riffler was taken for every batch of coal fed to the bench-scale unit. The analysis of this sample was used as the basis for the material balance calculations for each run. A typical analysis was shown in our previous paper (1).

Material Balance Calculations

As a result of using hydrogen as both the preheating gas and the quenching gas, the coal-derived, reaction products are very dilute in hydrogen. Liquor and aromatic oils (boiling above BTX) are condensed using conventional indirect heat exchange. The lighter products, methane through BTX, are measured using gas chromatography combined with calibrated flow meters. Rotameters, orifice meters and turbine meters are used to measure gas flow. Redundancy in flow metering was incorporated into the design of the bench-scale unit in order to minimize uncertainty in calculation of the gaseous product yields.

All solid and liquid products are weighed after every run. The reactors were examined after each run and found to be free of deposits. The recovery system is purged with high pressure steam after every run in order to remove any hydrocarbons or char that does not readily disengage from the tubing surfaces, valves, receivers, etc. These hydrocarbons and char are recovered from the steam condensate by extraction with ether and are included in the material balance.

A packed cryogenic condenser was developed to condense the residual BTX material from the ambient (70°F) hydrogen stream after G. C. analysis. The weighed amount of BTX liquids recovered from this condenser checked

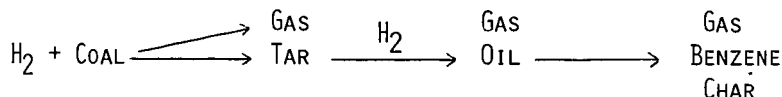
with the chromatographically-determined, BTX content of the gas stream entering the cryogenic condenser. Carbon balances of 90-105% are obtainable. Hydrogen consumption is calculated both 1) by difference of measured feed and product streams and 2) by elemental hydrogen balance between feed and product streams. Ash balances are 90-100%. The calculated oxygen balances are 100 \pm 10%. Nitrogen and sulfur balances are calculated by difference because of the very low quantities of these elements in lignite and lignite-derived, hydropyrolysis products. Gas yields are calculated based on coal fed to the reactor and corrected to 100% carbon balance.

CS-SRT Process Technology

Mechanism

The mechanisms involved in short residence time hydropyrolysis of coal are exceedingly complex. Coal consists of many types of structures each with different bond strengths. The initial coal conversion reactions are controlled by a thermal mechanism. Bond breaking will closely follow the rapid heating profile imposed on the coal. Weak bonds will rupture at the lower temperatures and decomposition products that are volatile will escape the particle under the driving force of a concentration or partial pressure gradient. As temperature is increased, stronger bonds will break and the volatiles will follow the same escape mechanism. These reactions are extremely rapid, of the order of milliseconds. Many of these devolatilized species are highly reactive and will undergo secondary decomposition reactions to form condensed and polymerized molecules generally classified as asphaltenes and char. However, these asphaltenes can be stabilized in the bulk phase and removed as single or double-ring aromatics by imposing the proper hydrogen partial pressure-temperature-residence time profile.

In terms of macro-reactions, the following simplified mechanism describes the vapor-phase, short residence time, hydrogenation process:



Although benzene is very stable, it too will decompose at elevated temperatures as shown by R. Graff and coworkers at CUNY (3). Extended residence times will also result in benzene decomposition as reported by Virk, et. al. (4). Normally, benzene decomposition in a hydrogen atmosphere would result in methane as the main decomposition product. However, it appears that in the case of benzene formation via short residence time coal hydropyrolysis, char particles catalyze the benzene-to-coke route of decomposition. Therefore, rapid quenching of the product vapors is necessary for the stabilization and recovery of aromatic liquids. The quantitative sequence of the in situ hydrogenation of tars to heavy oil to light aromatics is shown in Figure 2.

Coal Oxygen Reactions

Processing of lignite, because of its high oxygen content, provides some interesting observations about the mechanism of short residence time hydropyrolysis. The gaseous products obtained at very short residence time were rich in carbon dioxide while those obtained at longer residence time were depleted of essentially all carbon dioxide. This led to the conclusion that one of the initial reactions of short residence time

hydrogenation of lignite is the pyrolysis of carboxylic oxygen structures (decarboxylation). This agrees with the work reported by James Johnson at IGT (5). Once into the vapor phase, carbon dioxide hydrogenates to form carbon monoxide and water by the reverse water-gas shift reaction. Carbon monoxide will react further with hydrogen to form methane and additional water by the methanation reaction. These relationships are shown in Figures 3 and 4 which show the yields of water and methane respectively as a function of CO_2/CO ratio. The fact that these relationships were found in the 1/4-inch entrained-flow reactor and in the 1.1-inch, free-fall reactor, where the surface/volume ratio varied from about 50 to 500 in m^{-1} , tends to indicate that these vapor-phase reactions were not catalyzed by the SS 316 reactor wall. Equilibrium calculations of the reactor effluent also show that a) methane is approached from the carbon monoxide side of the steam-reforming reaction and b) carbon monoxide is approached from the carbon dioxide side of the water-gas shift reaction.

Coal Hydrogen Reactions

Another feature of the CS-SRT Process (as well as many other direct coal hydrogenation processes) is the utilization of coal hydrogen. Coal dehydrogenation increases continuously as hydrogenation severity and coal conversion are increased. During the initial pyrolysis stage of short residence time hydrogenation, the rate of coal dehydrogenation is increasing faster than hydrogen is being consumed. The rate of coal dehydrogenation appears to reach a maximum at about 30% carbon conversion as shown in Figure 5. At this point, approximately 50% of the hydrogen consumption is coming directly from the coal. This point also corresponds to the end of the pyrolysis mechanism and the initiation of the char hydrogenation stage. This is not surprising since 30% carbon conversion corresponds to 43% coal (MF basis) conversion which is identical to the ASTM volatile matter for lignite.

Furthermore, at carbon conversions greater than about 30%, hydrogen partial pressure has a pronounced effect on carbon conversion. The flux of volatiles escaping the coal particles decreases greatly above the 30% carbon conversion level. This results in increased hydrogen counterdiffusion into the char structure allowing more reaction between hydrogen and carbon sites. Higher hydrogen pressures greatly influence the diffusion of hydrogen into the particle. This is shown in Figure 6 and agrees with the work of James Gray and coworkers at PERC (6).

Rapid-Rate Carbon

One of the principal correlation tools of short residence time hydrolysis is the rate of conversion of rapid-rate carbon (7). The latter is a measure of the total theoretical amount of coal carbon that can be converted under "rapid" hydrogenation conditions. The rate itself is proportional to hydrogen partial pressure and the amount of rapid-rate carbon material remaining. Rapid-rate carbon is a function of coal type, particle size, temperature and pressure and is a difficult parameter to measure. Although this kinetic tool cannot be used as a predictive technique unless all of these functionalities are known, it can be used to demonstrate the relative effects of different hydrolysis data acquisition methods.

When processing a lignite at about 850°C , researchers (7) utilizing a captive sample technique for data acquisition reported a pseudo-rate constant of $50 \text{ atm}^{-1} \text{ hr}^{-1}$. Calculation of the pseudo-rate constant from free-fall reactor test data reported in the literature (8) results in a value of about $8 \text{ atm}^{-1} \text{ hr}^{-1}$ for lignite. Both sets of data were based on the assumption that the rapid-rate lignite carbon available for rapid hydrogenation is 100%. If the actual rapid-rate carbon for lignite is less

than 100%, then the respective pseudo-rate constants would both show corresponding increases.

The pseudo-rate constant for lignite at 850°C for the CS-SRT Process is 230 atm⁻¹ hr⁻¹, 5 to 30 times greater than that of the literature data, respectively. This CS-SRT pseudo-rate constant was based on data obtained with the turbulent, entrained-flow reactor equipped with a highly efficient, coaxial, coal-hydrogen mixing injector. Thus, it can be seen that in order to maximize carbon and thermal efficiencies in the conceptual CS-SRT reactor, it is imperative to utilize a reactor design based on data acquired from a bench-scale unit that has been operated under conditions closely scalable to the commercial reactor concept.

CS-SRT Conceptual Process

A conceptual plant design for producing 250 MM SCFD of pipeline gas and 6300 BPD of aromatic liquids has been developed. The design was based on data obtained when processing a North Dakota lignite in the bench-scale unit. Several alternative processing sequences have been examined but an optimization has not been undertaken. When more definitive test data become available, as would be generated in a pilot plant, a conceptual process optimization will be performed. The balance of this paper deals with the processing requirements about the reactor system including coal drying, coal feeding, SRT reactor designs and quench. Details of the downstream processing, hydrogen production and offsites are presented in another paper at this meeting.

Design Basis

After careful analysis of the CS-SRT Process data and patents, the following reactor design basis was established for the conceptual plant:

1. "Slow" heating of coal to the incipient devolatilization temperature in order to increase thermal efficiency;
2. "Rapid" heating of coal from the incipient devolatilization temperature to hydrogenation temperature in order to maximize rate of conversion;
3. Use of the reactant, hydrogen, as the heat carrier in order to minimize the number of streams to be handled;
4. Total residence time less than two seconds;
5. Rapid quenching of product vapors to prevent secondary decomposition reactions;
6. Recovery of high temperature heat from the reactor effluent.

Coal Drying

Lignite drying on the commercial-scale typical of an SNG plant has an energy requirement of about 16 billion BTU/day which equates to about 5.5% of the heating value (HHV) of the lignite. In conventional drying methods, this heat is supplied from hot combusted fuel gas. Efficient lignite drying is a relatively new technology and conventional processes have been applied to reducing the moisture from 35% to only about 10-15%. In the case of the CS-SRT Process, any moisture entering the reactor will be evaporated at the relatively high SRT reaction temperatures. This heat duty will of course come from the hot recycle hydrogen stream. Therefore, a design specification of 3% lignite moisture was set in order to improve the reactor heat balance. Reduction to 3% using conventional drying technology may prove difficult due to the long residence times required in gas conveyor-type dryers.

To circumvent these problems, coal drying in the CS-SRT Process will make use of the large quantity of low level heat available in the form of 60 psi steam. This steam is associated with the inefficiencies in the

many processing steps in all coal conversion plants. In the CS-SRT Process, it is believed that 60 psi steam will account for about 5% of the heating value of the coal fed to the plant.

Furthermore, advantage is taken of the boiling point depression of water as a result of the azeotrope formed between water and benzene (156°F, 1 atm, 30 mol% water, 70 mol% benzene). Benzene is chosen since it is a byproduct of the CS-SRT Process thereby minimizing the handling of additional chemicals. Accordingly, wet lignite will be pulverized and slurried with benzene as shown in Figure 7. The slurry will then be fed to an ebullated-bed contactor where it is heated by an inert recycle gas stream. The gas stream will obtain its heat by indirect heat exchange with the 60 psi steam source. Proper design of the ebullated-bed, which falls within Cities Service's LC-Fining technology, will result in the vaporization of the benzene-water azeotrope. Condensation of the azeotropic vapors results in the recovery of a relatively pure liquid benzene stream and a liquid water stream. The benzene is recycled and this water, generated internally within the process, will be available for plant use in the gasifiers or shift reactors. This water can account for approximately 40% of the total process water needed in the CS-SRT complex and can result in water savings of about 2000 acre-ft/yr.

The "dried" coal-benzene slurry will be drawn from the ebullated-bed with less than 3% water content. The slurry will now readily be available for downstream processing in the coal feed section.

Coal Feeding

Since the CS-SRT Process utilizes a pressurized gasifier, it has the same problems regarding the feeding of coal into a pressure vessel as do other pressurized gasification processes, such as BI-GAS, HYGAS, and SYNTHANE. One major difference and advantage is that the CS-SRT Process produces benzene, a solvent that can be used to maintain a pressure seal. Therefore, the low pressure, lignite-benzene slurry from the coal drying section will be pumped to system pressure with a positive displacement injection pump. The benzene will be subsequently flashed away from the lignite in a drier near its critical point. Benzene content of the dried coal should be extremely low as a result of the purging effect of the benzene-free, fluidizing gas.

Lignite with less than 3% water and at system pressure will then be delivered to a dense-phase transport system (see Figure 8) similar to that described by Rocketdyne Division of Rockwell International (9). Laboratory tests have shown lignite dried from a benzene slurry to be free-flowing. It is believed that the small quantity of benzene remaining in the coal pores will enter the reactor and be carried through along with the product gases without undergoing decomposition.

Reactor Design

Rapid heating of coal may be achieved by mixing hot recycle hydrogen with the dense-phase, transported coal in an injector assembly at the inlet of the reactor. With properly designed injectors, mixing is rapid on the order of a few milliseconds. Once mixed the coal and hydrogen will traverse the reactor upwards under conditions of fast fluidization to allow intimate contact between hydrogen and coal particles. The temperature and flowrates of the hot recycle gas and coal respectively will be set to allow the CS-SRT reactions to proceed adiabatically and exothermically. The recycle hydrogen/coal ratio is a critical parameter affecting not only the reactor heat balance and reactor kinetics but also the cost of downstream processing equipment.

As a result of the rapid heatup and short residence time features of the CS-SRT Process, extremely high coal throughputs are possible, of the order of 2000 lb/hr ft³ of reactor volume. For the case of a 250 MM SCFD

plant (and associated BTX production), only three reactors will be required, each capable of processing roughly 200 TPH coal through a 3 1/2 ft inside diameter.

Multi-Tube Reactor Concept

The combination of operating at a minimum hydrogen/coal recycle ratio under exothermic conditions can result in excessive temperature rises. This could result in loss of valuable benzene due to the adverse kinetics favoring benzene decomposition. Furthermore, since coal is being transported in dense-phase in a series of tubes to a multi-element injector assembly, a multi-tube reactor concept appears plausible.

Accordingly, a multi-tube, high pressure reactor with internal heat exchange is being developed. High pressure steam generation (1500 psi) can be used on the coolant side to maintain an isothermal tube wall. However, by using a coolant, such as a molten metal, heat can be absorbed non-isothermally while allowing control of the temperature profile within the reactor tube. This heat can also be used for high temperature heat exchange elsewhere in the process. Control of the reactor temperature profile will afford the plant operator the ability to vary product spectrum, conversion and turndown.

In one conceptual case, molten sodium is used to 1) control the reactor temperature profile and 2) limit tube wall temperature in contact with coal-hydrogen reactants to 1200°F. Other molten metal systems, such as molten lead or a 22% sodium/78% potassium eutectic, are also under consideration. The reactor design might consist of 12 alloy heat transfer tubes containing the reactants at system pressure. The tubes are immersed in the molten metal environment, also at system pressure, within a refractory-lined pressure vessel. The tubes will be about 12-inches in diameter and fit within the 3 1/2 ft. i.d. vessel. The major advantages of this reactor design are believed to be:

1. High coal throughput per volume of reactor;
2. Excellent turndown (8-100% full capacity);
3. Excellent reactor temperature control;
4. Overcomes heat transfer limitations of conventional pressure vessels;
5. Potential low risk to scaleup from pilot operations.

Molten Metal Heat Exchange

High thermal efficiency in the CS-SRT Process may be achieved by recovery of the high temperature heat of the reactor effluent. However, this has to be accomplished within the constraints of rapid quenching. This problem has been recognized and solved in the associated technology of ethylene production via steam cracking (10,11). In the latter reference, indications are that molten metal is used to rapidly quench steam cracker effluent while at the same time recovering high temperature heat. Quenching requirements for ethylene production are even more stringent than for the CS-SRT Process, of the order of 10-15 milliseconds compared to about 100 milliseconds.

A molten metal heat exchanger system has been conceptualized and applied to the CS-SRT Process. The bulk of the heat stored in the molten metal results from heat recovery in the reactor effluent quench exchanger. This heat can then be exchanged to provide the duties for recycle gas preheating, coal moisture removal and coal-benzene slurry drying. These total duties are equivalent to about 7.5 thermal efficiency points.

Our calculations have shown that this concept is feasible from both a process and a metallurgical standpoint. The concept utilizes molten metal technology already developed for the Clinch River breeder reactor.

Studies of the economics and optimization of this molten metal technology to the CS-SRT Process are continuing.

Conclusion

Bench-scale testing with a North Dakota lignite has shown that the CS-SRT Process scheme may be an attractive process for converting coal to pipeline quality gas, ethane and BTX liquids. Production of BTX liquids (and ethane feedstock for ethylene production) in the CS-SRT Process is a key factor in lowering the overall cost of services for producing SNG from coal. As a result of the rapid heatup, short residence time and rapid quench features of the process, a simple, high throughput reactor concept may be utilized without suffering loss in conversion and undesirable, secondary-decomposition reactions.

In order to maximize carbon and thermal efficiencies in the conceptual CS-SRT reactor, it is imperative to utilize a reactor design based on data acquired from a bench-scale unit that has been operated under conditions closely scalable to the commercial reactor concept.

Flowsheet studies of the CS-SRT Process have resulted in identifying several promising processing steps applicable to coal gasification:

1. Drying of coal using azeotropic benzene vaporization in an ebullated-bed contactor;
2. Multi-tube, high pressure, entrained-flow reactor concept with internal heat exchange and low scaleup ratio to commercial operation;
3. High temperature, heat recovery using molten metal heat exchange.

The next step of development of the CS-SRT Process would require about a 6-inch diameter, pilot-plant reactor capable of processing about 100 TPD coal. This pilot-plant would be one scaleup step away from a 12-inch diameter, single tube, commercial-type operation.

Acknowledgements

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Figure 1

COMPARISON OF REACTORS UTILIZED IN ENERGY RESEARCH LABORATORY

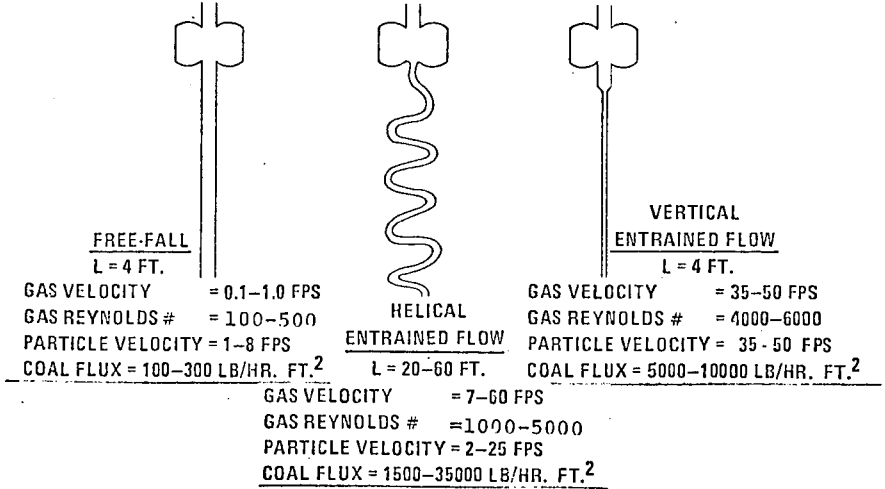


Figure 2

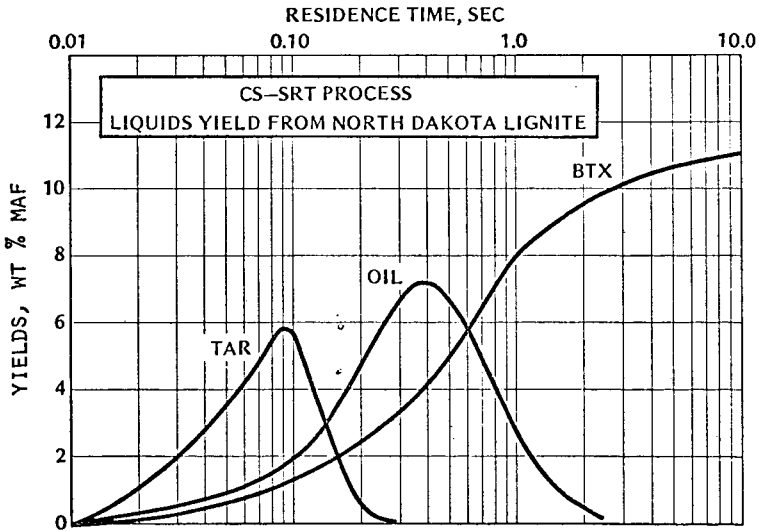


Figure 3

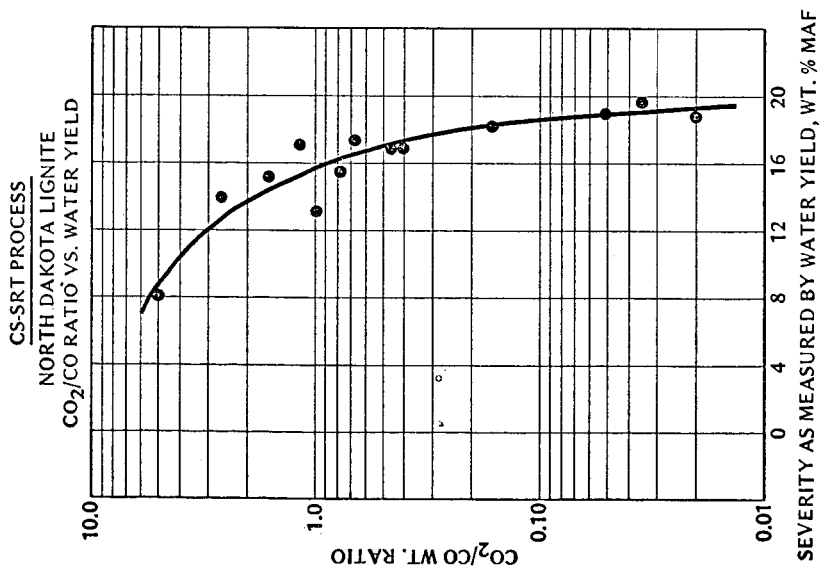


Figure 4

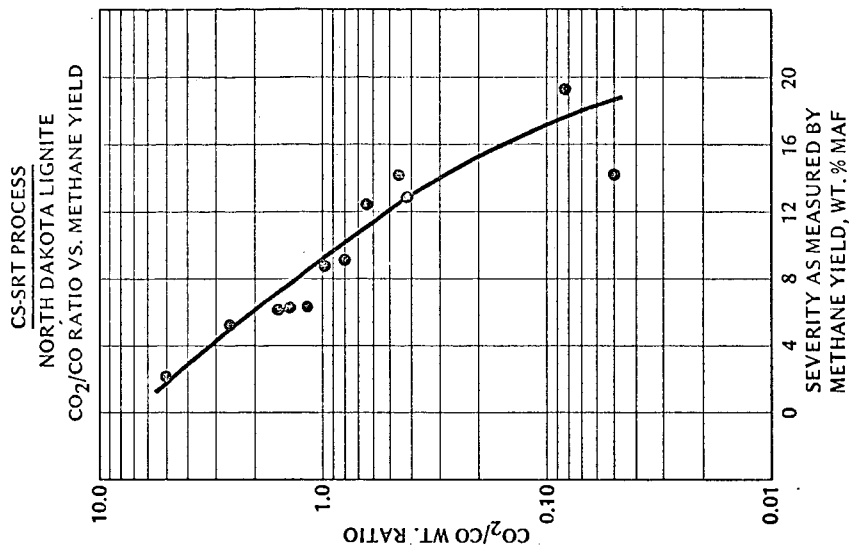


Figure 5

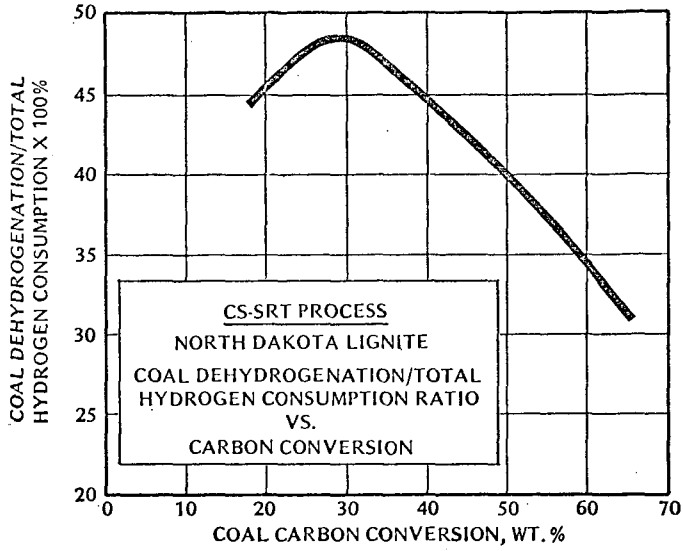


Figure 6

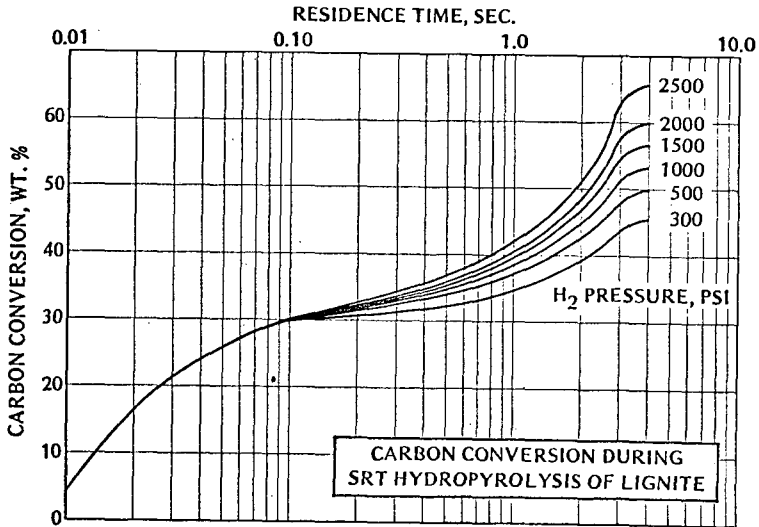


Figure 7
CS-SRT PROCESS
Coal Drying

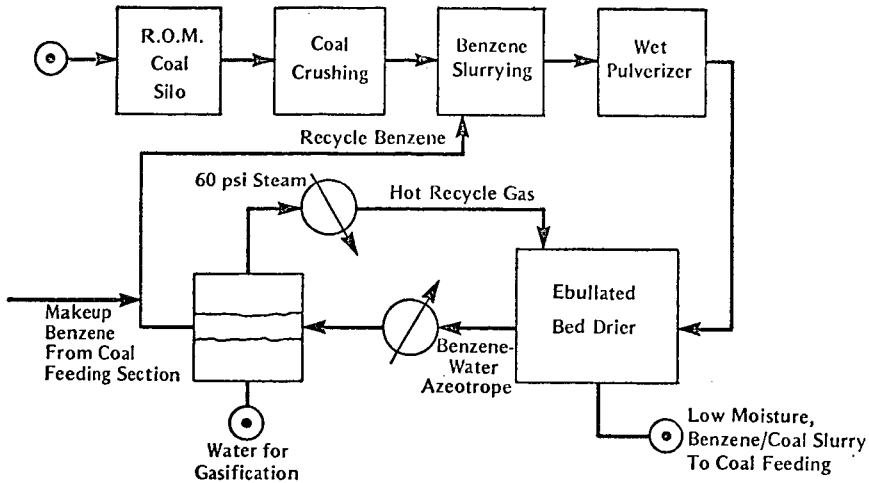
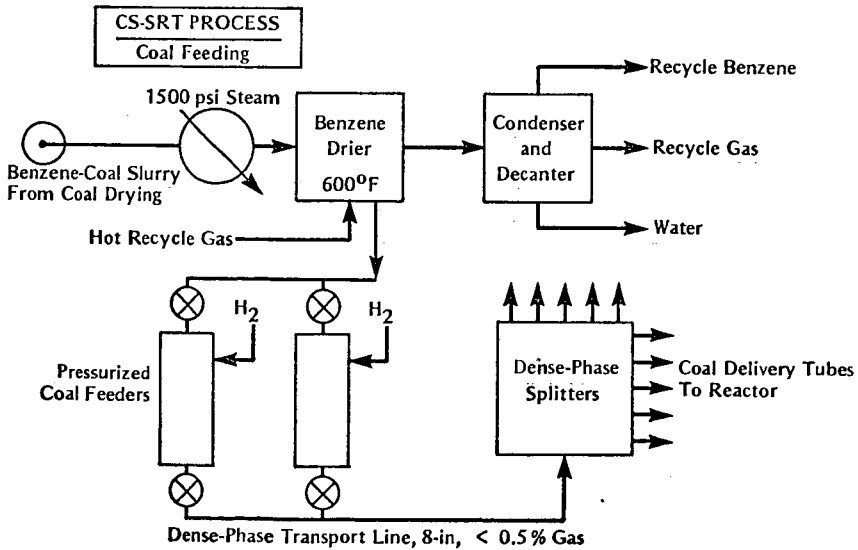


Figure 8



LIQUID FUELS FROM LOW Btu GAS: DILUTE PHASE FISCHER-TROPSCH SYNTHESIS

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Methods of producing liquid hydrocarbons from coal and other solid carbonaceous substrates may be roughly characterized as direct or indirect. Direct methods include various pyrolysis and catalytic hydrogenolysis techniques which are described in the literature (1,2). Indirect liquefaction methods first gasify the carbonaceous substrate to produce a synthesis gas which can then be converted into liquid hydrocarbons. The composition of the synthesis gas can be tailored to fit the needs of the subsequent hydrocarbon synthesis by proper choice of gasifying conditions, gasifying agents, CO-shift conversion and removal of undesirable constituents. Extensive investigations have been made on the use of synthesis gas composed of H_2 and CO for producing liquid hydrocarbons suitable as fuel and specialized chemicals. These reactions are categorically referred to as the Fischer-Tropsch synthesis although significant contributions have been made by a number of different investigators. The Fischer-Tropsch synthesis has been almost exclusively used with synthesis gas consisting of pure H_2 and CO of various stoichiometric ratios. In the following discussion a new application of this old technology for indirect coal liquefaction will be presented. This application consists of using synthesis gas diluted with inerts such as N_2 , CH_4 and CO_2 and is referred to as the Dilute Phase Fischer-Tropsch Synthesis.

CONVENTIONAL FISCHER-TROPSCH SYNTHESIS

The synthesis of hydrocarbons from H_2 and CO has been of considerable academic and practical interest since the discovery of methane synthesis by Sabatier in 1902. These simple reactants constitute the necessary building blocks for producing a large spectrum of essential organic chemicals and fuels. The synthesis has been reviewed several times over the past 50 years by Vannice, (3) Pichler, (4) Anderson, (5,6) Hofer, (7) and Storch et al. (8)

The diversity of the conventional Fischer-Tropsch synthesis is demonstrated in Figure 1. It is evident that a considerable spectrum of hydrocarbons can be obtained from pure H_2 and CO synthesis gas by proper choice of reaction conditions. The choice of catalyst is perhaps the most critical variable in the synthesis although pressure, temperature, H_2/CO ratio and space velocity or residence time are clearly important in determining product composition. Methane is the predominant product with nickel catalysts over a considerable pressure range at 200-300°C. Paraffins and olefins are produced over iron and cobalt catalysts in both the low (1 atm) and medium (5-20 atm) pressure synthesis at 185-300°C. At higher pressures (~ 100 atm) and temperatures ($\sim 400^\circ C$) the product over iron catalysts consists mostly of oxygenated hydrocarbons (Synthol). High molecular weight (100,000-200,000) paraffins are produced with ruthenium catalysts at 100-1000 atm pressure and 200-250°C. Methanol and higher alcohols are obtained from zinc oxide and alkali promoted zinc oxide catalysts, respectively, at high pressures (100-300 atm) and temperatures of 250-350°C. Using difficult to reduce metal oxide (ThO_2 , $Al_2O_3 + ZnO$) catalysts at high pressures and temperatures of 400-500°C aromatics, naphthenes and iso-paraffins are produced.

DULITE PHASE FISCHER-TROPSCH SYNTHESIS

In spite of the extensive work on the Fischer-Tropsch synthesis, little attention has been directed at employing diluted synthesis gas in the synthesis. Dilute synthesis gas is similar in composition to low Btu gas. It is obtained by gasifying carbonaceous material with air-steam or oxygen enriched air-steam mixtures. Contrary to gasification with O_2 -steam, the gas will contain moderate to high concentrations of nitrogen. The work that has been done with dilute synthesis gas, although sketchy, indicates that significant yields of liquid hydrocarbons can be obtained with conventional Fischer-Tropsch catalysts. Hall and Smith (9) examined the effect of nitrogen dilution on the F-T synthesis; the results are summarized in Table 1. As with non-diluted synthesis gas at constant temperature, the yield of product per volume of synthesis gas decreased with increasing space velocity. Furthermore, at constant space velocity replacement of synthesis gas with nitrogen results in an increase yield of liquids plus solids per cubic meter of $2H_2 + 1CO$. This is readily seen by comparing yields of the first with the second row at 1 atm and the fourth with the fifth row for the 10 atm synthesis. Gibson and Hall (10) studied the F-T synthesis with diluted synthesis gas over a Co-ThO₂-MgO-kieselguhr catalyst concluding that the dilution affected the rate of synthesis by reducing the partial pressure of reactants ($2H_2 + 1CO$). Dilution of the synthesis gas was also observed to increase the proportion of the low-boiling product. The observed variation in liquid product and CO conversion with dilution by these investigators is shown in Figure 2. Although the yield per unit volume of gas will be less in the dilute phase F-T synthesis, the product composition is likely to be similar to conventional F-T synthesis under similar operating conditions. A typical product composition over a cobalt catalyst in the conventional F-T synthesis is shown in Figure 3.

UTILITY OF DILUTE PHASE F-T SYNTHESIS

In the preceding section we have shown that dilute phase F-T synthesis can be used to obtain liquid hydrocarbons from synthesis gas ($H_2 + CO$) that is highly diluted with N_2 , CH_4 and CO_2 . With this capability, the dilute phase Fischer-Tropsch synthesis is viewed as an add-on facility where liquid hydrocarbons are obtained as a byproduct from low Btu gas. Although any low Btu gas could be used in the synthesis, in situ generated low Btu coal gas has been used to carry out preliminary engineering and economic feasibility studies. The conceptualized process is shown in Figure 4. Stepwise, coal is first gasified in situ with air. The in situ generated gas is then subjected to a gas clean-up where H_2S , H_2O , CO_2 and dust are removed. This is followed by dilute phase F-T synthesis and product fractionation to obtain the liquid hydrocarbon byproduct from the low Btu gas. Material balance calculations based on situ coal gasification data by Brandenburg et al. (13) and the dilute phase F-T synthesis data of Gibson and Hall (10) for a 50,000 bbl/da plant are shown in Table 2. Hence, 86×10^8 SCF/da of air is used for the in situ gasification of 2.16×10^5 ton/da of coal. The removal of impurities provides 147.7×10^8 SCF/da of dry, dilute, low Btu (152 Btu/SCF) synthesis gas. Dilute phase F-T synthesis yields 50,000 bbl/da of liquid byproduct with the primary product being 121.0×10^8 SCF/da of low Btu (148 Btu/SCF) fuel gas. The liquid yield amounts to 1/4 bbl/ton of in situ coal gasified. The liquid would be sulfur and nitrogen free and estimated to contain up to 25% olefins with 57% boiling below 150°C and 34% boiling between 150-270°C.

TABLE 1. Effect of Diluted Synthesis Gas on F-T Synthesis⁽⁹⁾
(Co-ThO₂-MgO-kieselguhr catalyst)

Temperature °C	Pressure atm	Feed Gas Composition					Space Velocity hr ⁻¹	Yield of Liquids and Solids	
		volume %						g/m ³	
		H ₂	CO	N ₂	CO ₂	CH ₄		Feed Gas	2H ₂ +1CO
193	1	68.7	30.0	1.0	--	0.3	184	109.2	110
193	1	58.2	24.2	16.3	--	1.3	91	106	127
193	1	44.4	26.7	28.8	--	0.1	133	87.8	123
192	10	62.0	31.5	4.0	2.5	--	131	106	113
193	10	41.8	20.0	35.1	3.1	--	138	112	120

TABLE 2. Process Stream Compositions

Process Stream #	Description	Composition, Vol. or Wt. %	Flow
1	Air	80 N ₂ , 20 O ₂	86 x 10 ⁸ SCF/da
2	Coal	6.3 H ₂ O, 18.4 ash, 0.5 S, 4.6 H, 55.3 C, 0.7 N, 14.2 O	2.16 x 10 ⁵ tn/da
3	Low Btu gas (102 Btu/SCF)	26.9 H ₂ O, 12.2 H ₂ , 6.1 CO, 0.6 O ₂ , 36.5 H ₂ , 3.3 CH ₄ , 0.4 C ₂ , 14.0 CO ₂ , 0.05 H ₂ S	253 x 10 ⁸ SCF/da
4	Scrubbed gas impurities	? dust, 0.12 H ₂ S, 64.8 H ₂ O, 35.0 CO ₂	105 x 10 ⁸ SCF/da
5	Dry dilute phase synthesis gas (152 Btu/SCF)	20.7 H ₂ , 10.3 CO, 1.0 O ₂ , 61.8 N ₂ , 5.6 CH ₄ , 0.7 C ₂	147.7 x 10 ⁸ SCF/da
6	Synthesis product and unconverted gas	0.35 C ₁ H _{2.4} (estm.), 8.1 H ₂ , 4.6 CO, 69.6 N ₂ , 6.5 CH ₄ , 1.7 C ₂ , 8.8 H ₂ O, 0.2 C ₃ , 0.2 C ₄	133.6 x 10 ⁸ SCF/da
7	Liquid hydrocarbon	C ₁ H _{2.4} (estm.), density = 0.85 g/cm ³ (estm.)	5 x 10 ⁴ bbl/da
8	Low Btu gas (148 Btu/SCF)	8.9 H ₂ , 5.1 CO, 76.5 N ₂ , 7.1 CH ₄ , 1.9 C ₂ , 0.2 C ₃ , 0.2 C ₄	121.0 x 10 ⁸ SCF/da
9	Water	100 H ₂ O	11.9 x 10 ⁸ SCF/da

PROCESS ECONOMICS

Preliminary economics have been calculated for the conceptualized dilute phase F-T synthesis coupled with in situ coal gasification. These calculations, based on the material balance shown in Table 2 and certain necessary assumptions, indicate that high quality sulfur-free liquid fuels can be produced for as little as \$13/bbl. This assumes use of an iron catalyst costing \$0.10/lb, low Btu gas from in-situ gasification at \$0.43/MM Btu as described by A. D. Little, (11) and a 10% return on capital investment.

The cost of the liquid fuel produced is highly dependent on capital investment and raw materials costs so any increase in space velocity or increase in yield would significantly reduce the cost of the liquid product. Likewise an increase in the cost of raw materials (low Btu gas, catalyst, etc.) would increase the liquid fuel cost. The influence of these variables on liquid fuel cost is shown in Table 3. These liquid fuel costs are based on a 10% return on investment and a six month catalyst life. Two different space velocities, 1000 hr⁻¹ and 2000 hr⁻¹ have been used in the calculations. The parenthesized cost figures are for the 2000 hr⁻¹ space velocity operations. The economic incentive for operating at higher space velocities or larger through-puts is clearly evident from the data in Table 3. It is also evident from the tabulated data that as the catalyst cost increases so does the benefit from operating at higher space velocities.

TABLE 3. Liquid Fuel Cost

	Product Cost \$/bbl		
	Cost of Low Btu Gas \$/MM Btu		
<u>Catalyst Cost</u>	<u>\$0.43</u>	<u>\$0.75</u>	<u>\$1.00</u>
\$0.10/lb	13 (12)	18 (16)	21 (20)
\$1.00/lb	23 (16)	28 (21)	32 (25)

Total capital investment for a dilute phase F-T facility producing 50,000 bbl/da of liquid fuels would be \$300-400 million in 1976 dollars. The major processing unit required is the synthesis reactor. Cost of the reactors is about one third of the total capital investment because of the large heat transfer area required to maintain proper control of reaction temperature. The heat of reaction will be recovered by generating steam to be used as process feed, in steam driven pumps and compressors, and for electrical power drives. Types of reactors that could be used range from a simple vertical boiler with catalyst packed in the tubes to a design where the catalyst is sprayed on the heat transfer surface. (12)

CONCLUSIONS

The dilute phase F-T synthesis is a new application of old technology. It provides a means of obtaining significant yields of liquid hydrocarbon byproduct from low Btu gas. Since the low Btu gas is obtained by gasifying with air in place of oxygen the need for separating O_2 and N_2 is eliminated. Engineering and economic evaluations were made of dilute phase F-T synthesis coupled with in situ coal gasification. The complex was scaled to produce 50,000 bbl/da of liquid byproduct and 121×10^8 SCF/da low Btu (148 Btu/SCF) fuel gas primary product. Considering such economic factors as return on capital investment, catalyst cost and life, space velocity and low Btu gas cost, the liquid product cost was estimated to be between \$13-35 bbl. The most significant contributions to the product cost were identified as return on investment, capital cost and catalyst cost. Technical indications are good that significant reductions can be made in the latter two areas which will improve the overall economics.

ACKNOWLEDGEMENT

This work was supported by the Office of Molecular, Mathematical and Geo-Sciences, Division of Physical Research, U. S. Energy Research and Development Administration.

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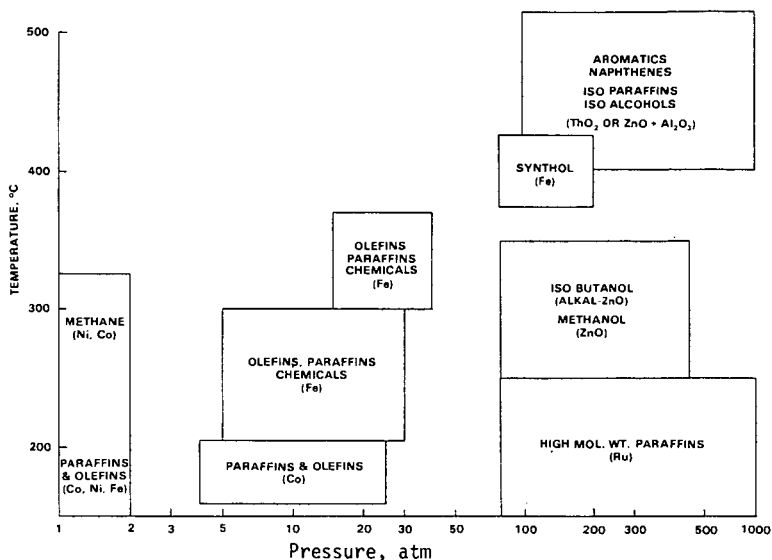


FIGURE 1. Pressure-Temperature Regions for Synthesis with CO and H₂

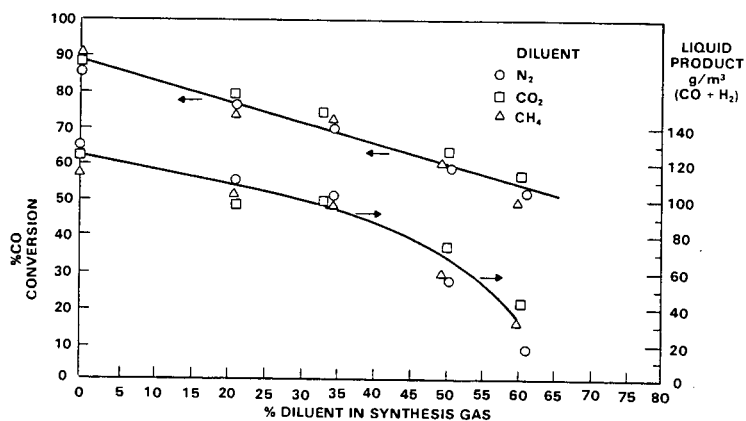


FIGURE 2. Dilute Phase Fischer-Tropsch Synthesis (Co-Catalyst, 1 atm, 185°C, 2H₂/lCO, 900 hr⁻¹ Space Velocity)

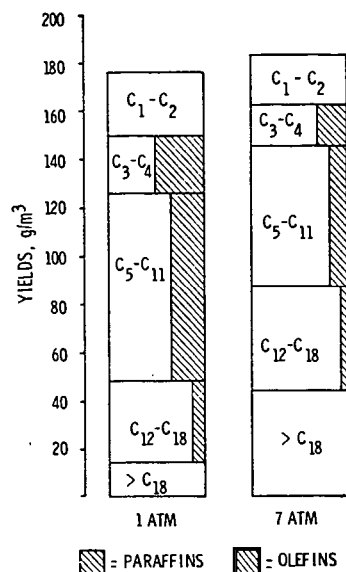


FIGURE 3. Typical yields with Co-ThO₂-MgO-kieselguhr catalyst at 1 atm. and 7 atm.

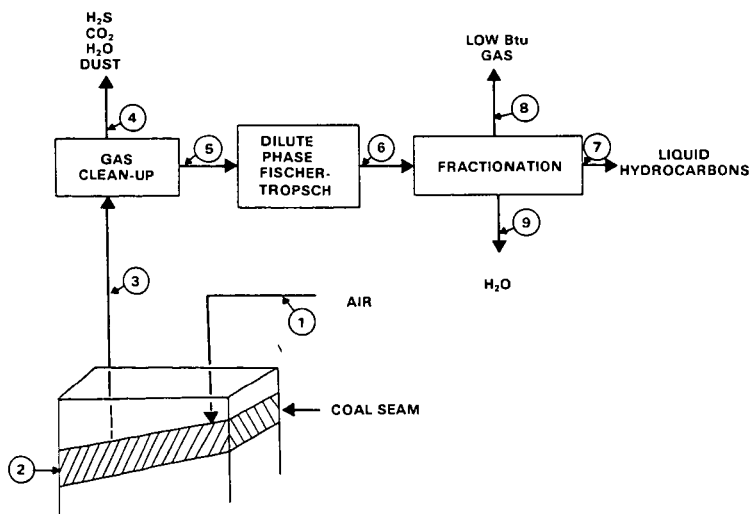


FIGURE 4. Dilute Phase Fischer-Tropsch Synthesis for In Situ Coal Gasification

ECONOMIC COMPARISON OF COAL FEEDING SYSTEMS IN COAL GASIFICATION--LOCK HOPPER VS SLURRY

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INTRODUCTION

Coal feeding to the various pressurized coal gasification processes presents certain technical problems that will have to be solved. Two feeding systems being considered are the lock hopper and coal slurry. The coal slurry system has the advantage of being simple to operate with a good reliability, but erosion problems in the circulating pumps and injection valves will have to be solved. The lock hopper system has the advantage of feeding a dry coal without solvent dilution, but will also have erosion problems in the valves and will require a complex cycle control system.

An economic comparison was made of the two feeding systems based on the Bituminous Coal Research's Bi-Gas process using a Western Kentucky No. 11 coal, assuming the two systems are technically feasible. The estimates are based on January 1976 cost indexes. Average selling prices of the gas were determined by using discounted cash flow rates of 12, 15, and 20 percent at various coal costs. No inflation factors are considered for the life of the plant. Pollution abatement considerations have been incorporated. Some of the economic and technical details of the Bi-Gas process are included.

Lock Hopper System

The Bi-Gas process is a two-stage coal gasification system to convert bituminous and subbituminous coal to a high-Btu pipeline gas. Figure 1 is a flowsheet of the process (1) and includes the following steps:

1. Coal preparation, which includes crushing, screening, sizing of the run-of-mine coal, and conveying the sized coal to the lock hopper feeding system (2) (not shown in figure 1).
2. In the upper section of the gasifier, coal contacts the hot synthesis gas produced in the lower section and is partially converted to methane and more synthesis gas. The entrained char in the raw product gas is separated and recycled to the lower section, where it is gasified to produce synthesis gas and heat required in the upper section.
3. Shift conversion of the synthesis gas to $H_2:CO$ ratio of 3.1:1.
4. The hot-carbonate purification system reduces the CO_2 content of the shifted gas to 0.5 percent and removes essentially all the H_2S and COS . Zinc oxide towers are provided for residual sulfur cleanup.
5. The heating value of the clean gas is increased to 946 Btu by reacting hydrogen with 99.8 percent of the CO in the presence of a nickel catalyst to form methane and water.
6. After gas cooling, the moisture content of the product is reduced to 7 pounds per million standard cubic feet.

The gasifiers are designed to operate at an outlet pressure of 1,160 psig and at maximum temperatures of 1,700° F and 2,700° F for the upper and lower sections, respectively. The thermal efficiency of the plant, using a Western Kentucky No. 11 coal, is 65.3 percent, based on a coal heating value of 13,070 Btu per pound.

Slurry Feeding System

The pulverized coal is mixed with hot condensate to produce a 50-50 weight-percent coal-water slurry and then raised to the system pressure of approximately 1,200 psig with triplex reciprocating pumps. After being preheated to 430° F, the coal slurry is dried in a spray dryer with gasifier product gas that vaporizes the water. The coal and humidified gas at 660° F flow to a cycle separator where the coal drops into a coal hopper and is pneumatically fed to the upper section of the gasifier. This feeding system (3) is illustrated in figure 2. The rest of the integrated plant, (4) shown in figure 3, is the same as described in the lock hopper system except for some changes in the shift conversion, gasification, and heat recovery units owing to shifting of heat loads. The thermal efficiency of the plant is the same as that of the lock hopper system--65.3 percent.

Capital Investment

An economic evaluation was made of an integrated plant sized and designed to produce 250 million scf of high-Btu gas (946 Btu/scf) by two-stage gasification of Western Kentucky No. 11 coal followed by shift conversion, purification, methanation, and pollution control. The total investment is estimated to be \$533.3 million for the lock hopper system, or \$92.4 million higher than for the coal slurry system.

Table 1 is a capital requirement comparison of the two feeding systems. Detailed cost summaries of the major processing units are not included, but the costs of the individual units are listed. General facilities include administrative buildings, shops, warehouses, railroad spurs, rolling stock, roads, waste water treatment, and fences. The costs of steam and power distribution, cooling water towers, plant and instrument air, fire protection, and sanitary water are included in plant utilities.

Operating Cost

Table 2 presents the estimated operating cost comparison for the lock hopper and coal slurry feeding systems. An assumed 90-percent operating factor allows 35 days for downtime, two 10-day shutdowns for equipment inspection and maintenance, and 15 days for unscheduled operational interruptions. With labor at \$6 per hour, payroll overhead at 30 percent of payroll, and depreciation at 5 percent of the subtotal for depreciation, allowing credit for sulfur recovered at \$25 per ton, and with the cost of coal as a variable, the following operating costs are derived:

Cost of coal per ton	Per year, MM		Per MMBtu	
	Lock hopper system	Slurry feeding	Lock hopper system	Slurry feeding
\$11	\$128.0	\$115.8	\$1.64	\$1.48
13	137.1	125.0	1.75	1.60
15	146.3	134.1	1.87	1.72

Based on a 330-day operating year for the plant and allowing credit for sulfur produced, with coal costs and discounted cash flow (DCF) rates as parameters, the average selling prices of the gas per Mscf and per MMBtu for the two systems are shown in the following table:

Coal cost, dollar per ton	Gas selling price											
	Dollars per Mscf						Dollars per MMBtu					
	12-pct DCF		15-pct DCF		20-pct DCF		12-pct DCF		15-pct DCF		20-pct DCF	
	Lock hop- per	Coal slurry	Lock hop- per	Coal slurry	Lock hop- per	Coal slurry	Lock hop- per	Coal slurry	Lock hop- per	Coal slurry	Lock hop- per	Coal slurry
11	2.61	2.28	2.97	2.58	3.64	3.13	2.76	2.41	3.14	2.73	3.85	3.31
13	2.72	2.39	3.08	2.69	3.75	3.24	2.88	2.53	3.26	2.84	3.96	3.42
15	2.83	2.50	3.19	2.80	3.86	3.35	2.99	2.64	3.37	2.96	4.08	3.54

The DCF computer program takes into account the capital expenditure, based on 100-percent equity, prior to startup so that the interest during construction is deleted from the capital requirement. Provisions are made for recovery of the working capital in the 20th year.

Unit Cost Summary

The selling price used to determine the high-cost elements in the process was based on a 15-percent DCF for a 20-year project life, with coal at \$13 per ton. A breakdown of the cost elements for the two systems is shown in table 3.

SUMMARY

As noted in table 1, the lock hopper feeding total investment is \$92.4 million, or 21 percent higher than the slurry feeding system investment. Approximately \$69 million is attributed to difference in feeding systems.

The operating cost of the lock hopper system is about 10 percent higher than that of the slurry feeding system. Increases in maintenance, overhead, and indirect and fixed costs, which are directly related to the capital investment, represent the main difference.

The average selling price was based on three coal costs (\$11, \$13, and \$15 per ton) and three DCF rates of return (12, 15, and 20 percent). Over this range the selling price for the lock hopper system increases from \$0.33 to \$0.51 per Mscf of product, or \$0.35 to \$0.54 per MMBtu. This increase is approximately 15 percent.

CONCLUSION

Results of this study indicate the slurry feeding system is more economical than the lock hopper system, when used to feed a high-pressure (1,200 psig) two-stage gasifier in the Bi-Gas process. It must be kept in mind, however, that this study was conducted under the assumption that the technical problems for both the coal slurry and lock hopper systems have been solved.

REFERENCE

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TABLE 1. - Capital requirements, comparison of
lock hopper system with slurry feeding system

Unit	Lock hopper system	Slurry feeding system	Difference
Coal preparation.....	\$14,295,400	\$14,295,400	0
Slurry preparation.....	-	16,435,000	\$+68,743,000
Lock hopper system.....	85,178,000	-	0
Gasification.....	20,698,500	20,698,500	0
Heat recovery No. 1.....	3,402,400	-	+3,402,400
Dust removal.....	2,341,100	2,341,100	0
Shift conversion.....	10,033,200	7,700,000	+2,333,200
Heat recovery No. 2 or 1	8,143,900	23,392,600	-15,248,700
Purification.....	75,713,300	75,281,100	+432,200
Methanation.....	18,782,900	18,782,900	0
Heat recovery No. 3 or 2	21,718,000	21,718,000	0
Drying.....	873,500	873,500	0
Oxygen plant.....	51,975,000	51,975,000	0
Sulfur recovery.....	2,312,500	2,312,500	0
Flue gas processing.....	16,033,300	16,033,300	0
Steam plant.....	22,822,500	20,910,000	+1,912,500
Plant facilities.....	26,574,200	21,956,200	+4,618,000
Plant utilities.....	38,089,700	31,470,500	+6,619,200
Total construction.....	418,987,400	346,175,600	+72,811,800
Initial catalyst requirements.....	2,559,800	2,348,100	+211,700
Total plant cost.....	421,547,200	348,523,700	+73,023,500
Interest during construction.....	63,232,000	52,278,600	+10,953,400
Subtotal for depreciation.....	484,779,200	400,802,300	+83,976,900
Working capital.....	48,477,900	40,080,200	+8,397,700
Total investment...	533,257,100	440,882,500	+92,374,600

TABLE 2. - Annual operating cost,
comparison of lock hopper system with slurry feeding system

Cost item	Lock hopper system	Slurry feeding system	Difference
Direct cost:			
Raw materials:			
Coal at \$11/ton.....	\$50,294,400	\$50,294,400	0
Raw water.....	1,176,100	1,211,800	\$-35,700
Catalyst and chemicals.	2,517,300	2,504,200	+13,100
Methane.....	455,600	455,600	0
Subtotal.....	54,443,400	54,466,000	-22,600
Direct labor.....	2,470,300	2,522,900	-52,600
Direct labor supervision..	370,500	378,400	-7,900
Subtotal.....	2,840,800	2,901,300	-60,500
Maintenance labor.....	7,815,000	6,465,000	+1,350,000
Maintenance supervision..	1,563,000	1,293,000	+270,000
Maintenance material and contracts.....	11,722,500	9,697,500	+2,025,000
Subtotal.....	21,100,500	17,455,500	+3,645,000
Payroll overhead.....	3,665,600	3,197,800	+467,800
Operating supplies.....	4,220,100	3,491,100	+729,000
Total direct cost.	86,270,400	81,511,700	+4,758,700
Indirect cost.....	11,264,600	9,539,200	+1,725,400
Fixed cost:			
Taxes and insurance.....	8,430,900	6,970,500	+1,460,400
Depreciation.....	24,238,900	20,040,100	+4,198,800
Total, before credit.	130,204,800	118,061,500	+12,143,300
Sulfur credit.....	2,226,700	2,226,700	0
Operating cost, after credit	127,978,100	115,834,800	+12,143,300

TABLE 3. - Unit cost comparison

Process unit	Cost per Mscf of product	
	Lock hopper system	Slurry feeding system
Coal preparation	\$0.13	\$0.13
Gasification	1.32	1.41
Lock hopper system50	-
Slurry preparation.....	-	.16
Dust removal02	.02
Shift conversion18	.05
Purification64	.63
Methanation17	.17
Drying01	.01
Sulfur recovery	-	-
Flue gas processing11	.11
Total	3.08	2.69

Basis: Coal at \$13/ton - 15% DCF.

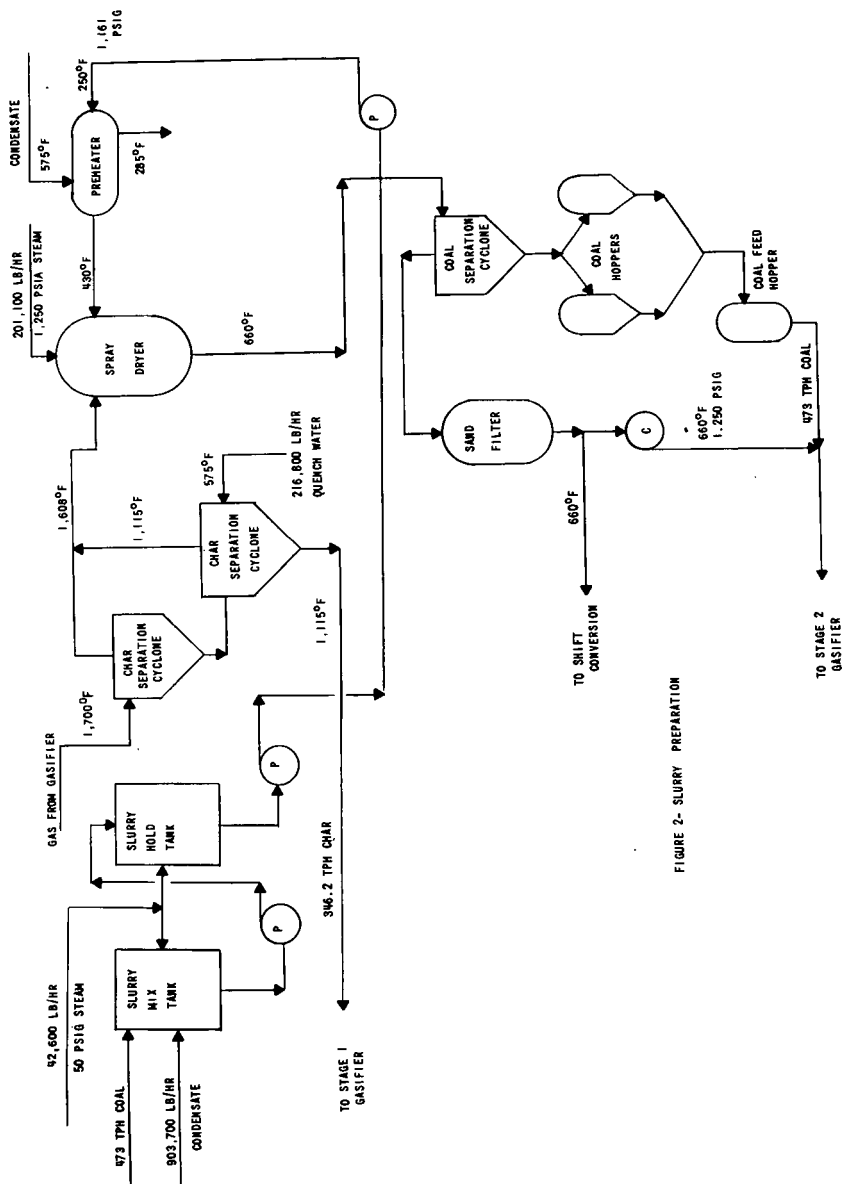


FIGURE 2- SLURRY PREPARATION

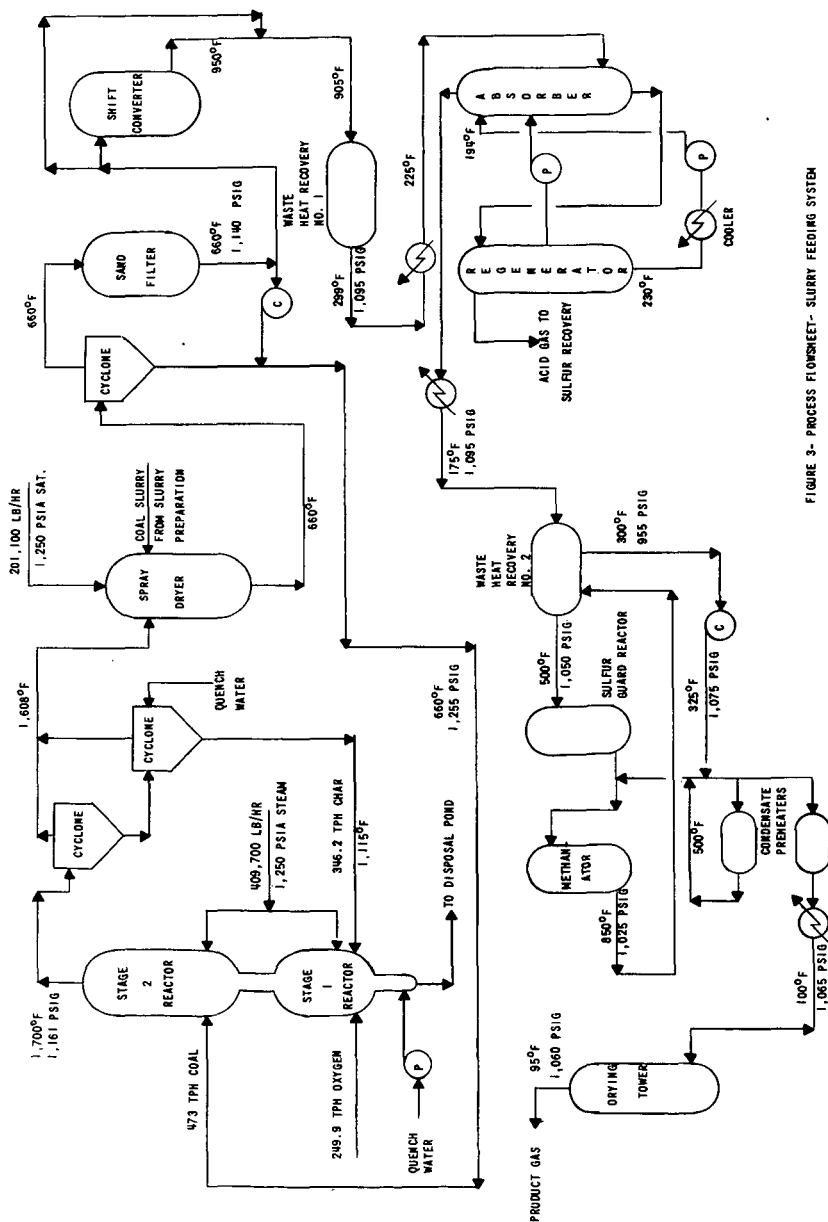


FIGURE 3- PROCESS FLOWSHEET- SLURRY FEEDING SYSTEM

Coal Desulfurization During Gaseous Treatment

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Sulfur removal from coal during treatment in gaseous atmospheres at elevated temperatures is of interest both because such a treatment might serve as a basis for a coal desulfurization process and because knowledge of the fate of sulfur under such conditions has application to the behavior of coal during carbonization or gasification. In the work reported here, desulfurization of both a raw and deashed Iowa coal were investigated in three different gases, nitrogen, hydrogen and a 6% oxygen-94% nitrogen gas mixture. With each gas, both the temperature and holding time at temperature were varied. With hydrogen and nitrogen, the temperature was varied between 300 and 700°C. The temperature range used with the oxygen-nitrogen mixture was only from 350 to 455°C due to the large weight loss at higher temperatures caused by combustion of the sample. Holding time at temperature was varied from 0 to 60 min.

EXPERIMENTAL

The investigation was carried out in a Rigaku CN8001 H type thermal analyzer which included a thermal gravimetric analyzer, a preprogrammed heating unit and temperature control system, and a data recording unit. To carry out a run, about 300mg of coal (<125 μ) was placed on the sample pan of the thermal analyzer and heated to the desired temperature using a heating rate of 20°C/min while maintaining a flow of gas over the sample. After holding the sample at temperature for the desired period of time, the gas flow was switched to nitrogen and the reaction chamber was cooled with the initial cooling rate being on the order of 250°C/min. The sample weight was recorded continuously, and at the end of the run the total and organic sulfur contents of the treated coal were determined.

The coal came from the Jude mine, Mahaska County, Iowa, and is a high-volatile-C bituminous coal. The raw coal contained 3.25% inorganic sulfur and 3.04% organic sulfur (moisture-free basis). The deashed Jude coal was prepared by twice floating the raw coal in a heavy liquid medium having a specific gravity of 1.3. It contained 4.74% organic sulfur, but only 0.69% inorganic sulfur. Thus, the results of experiments using the deashed coal manifested largely the behavior of the organic sulfur compounds in the coal. The analyses of the raw and deashed coal are shown in Table 1.

Table 1. Proximate analyses of coal samples

Proximate Analysis	Raw Coal	Deashed Coal
Moisture	1.22%	2.21%
Ash	18.80%	5.54%
Volatile matter	39.68%	45.72%
Fixed carbon	40.30%	46.53%

*Currently at Allis-Chalmers, Milwaukee, Wisconsin.

RESULTS AND DISCUSSION

Effect of Temperature and Holding Time

The effect of temperature upon sulfur removal in each of the three gases was investigated by heating the coal sample to the desired temperature using a heating rate of 20°C/min. followed by immediate cooling. In nitrogen, the amount of sulfur in all forms except for the organic sulfur in the raw coal tended to decrease with increasing temperature, with the decrease being greater for the deashed coal. At 700°C, the total sulfur content of the raw coal and the total and organic contents of the deashed coal were 5.60, 3.27 and 3.05 wt.%, respectively. On the other hand, the organic sulfur content of the raw coal increased from 3.04 to 4.82 wt.% as the temperature was increased from room temperature to 700°C. Since the raw coal contained a substantial amount of inorganic sulfur, this result indicated that inorganic sulfur was being transformed into organic sulfur during the heating process.

The various sulfur contents of the coal samples were not greatly affected when the coal was held at constant temperature. At lower temperatures, the sulfur contents tended to decrease slightly, while they tended to increase when held at higher temperatures.

Fig. 1 shows the effect of temperature on the total and organic sulfur content of the raw and deashed coal when treated with hydrogen. Again, except for the organic sulfur content of the raw coal which increased slightly, the various sulfur contents all decreased with increasing temperature. When the coal was held at temperature for extended periods, there was little change in the sulfur content at temperatures below 600°C. However, at higher temperatures the sulfur content decreased significantly when the sample was held at temperature in hydrogen.

The effectiveness of hydrogen for preferential removal of the organic sulfur can be determined by comparing the differences in the amount of organic sulfur removed during treatment with nitrogen and hydrogen. Any desulfurization in nitrogen is due to the evolution of volatile compounds containing sulfur. Therefore, no matter what the temperature or treatment time, a unique relationship should exist between the organic sulfur removed and the weight loss. The data collected with deashed Jude coal did indeed exhibit such a relationship (Fig. 2).

In hydrogen, sulfur is removed either by devolatilization, as in nitrogen, or by reaction of the hydrogen with sulfur in the organic matrix to form hydrogen sulfide. The effectiveness of hydrogenation for desulfurization can be judged by comparing the organic sulfur removed versus weight loss curves in hydrogen and nitrogen (Fig. 2). Up to a weight loss of about 40% (point A, Fig. 2), the organic sulfur loss curves in the two gases were nearly the same. However, at higher weight losses, the organic sulfur removed in hydrogen increased rapidly with only a slight increase in weight loss indicating that the sulfur-hydrogen reaction was taking place. All the experimental points at weight losses greater than that shown by point A were taken at 600 and 700°C. Reaction of hydrogen with the sulfur in coal appears therefore to be significant only at temperatures above 600°C. At lower temperatures, desulfurization in both hydrogen and nitrogen occur only due to removal of volatile, sulfur-containing compounds. The onset of the hydrogen-sulfur reaction accounts for the reduction of sulfur content with increased holding time at temperatures above 600°C.

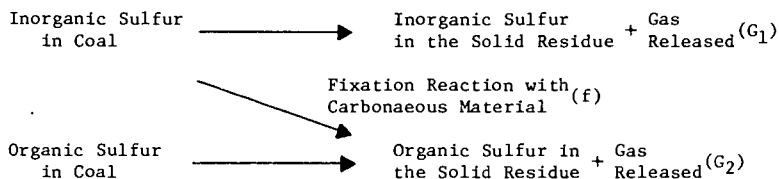
The final gas tested was a mixture of nitrogen with 6% oxygen. The effect on the sulfur content of treatment in the range from 350 to 455°C was slight, with a small decrease in the amount of all forms of sulfur occurring if the coal was held at a temperature in this range. However, the amount of sulfur in the coal decreased fairly substantially as the coal was heated from room temperature to

350°C in an oxidizing atmosphere with the sulfur contents of the two coals at 350°C being as follows:

	Sulfur Content, wt. %	
	Total	Organic
Raw coal	4.77	2.58
Deashed coal	3.87	3.39

Transformation of Sulfur

As previously noted, the results of the experiments indicated that some of the inorganic sulfur was trapped by the organic portion of the coal during the heating process. This same phenomenon has been observed by Snow (1) and Cernic-Simic (2). In an attempt to estimate the fraction of inorganic sulfur released as gas (G_1) or transformed into organic sulfur (f) and the fraction of original organic sulfur released as gas (G_2), the following reaction scheme was postulated.



By assuming that this scheme applied to both the raw and deashed coal, G_1 , G_2 and f were estimated from sulfur material balances. Details of the calculations are available elsewhere (3).

The fixation reaction, characterized by f , started at 300°C in nitrogen and increased rapidly between 400 and 500°C with f reaching 0.7 at 700°C. G_1 was generally small at all temperatures, being between 0 and 0.1. G_2 increased rapidly between 300 and 500°C, and then started to level off between 600 and 700°C. About 70% of the original organic sulfur was released as gaseous compounds at 700°C. In nitrogen, the effect on G_1 , G_2 and f of holding time when the sample was kept at constant temperature was small.

The fraction of inorganic sulfur transformed into organic sulfur (f) was larger in hydrogen than in nitrogen at low temperatures (Fig. 3). However, f remained constant at temperatures above 400°C. G_1 was again small, while G_2 increased with increasing temperature as it had in nitrogen. f and G_2 increased slowly when the samples were held at 400°C, while G_1 remained relatively constant. At 700°C, G_1 and G_2 both increased slightly with holding time, but f decreased from 0.5 to about 0.2 when the holding time was varied from 0 to 60 min. Apparently the H_2S formed from the organic sulfur reacted with the iron to produce FeS .

The fraction of inorganic sulfur transformed into organic sulfur (f) in the oxidizing atmosphere was comparable to that in nitrogen at the same temperature. It increased rapidly from about 0.1 at 350°C to 0.36 at 450°C. The fraction of inorganic sulfur released into the gas phase (G_1) was much larger than in nitrogen or hydrogen since the reaction of pyrite with oxygen is thermodynamically more favorable than the reaction of pyrite with hydrogen. G_1 was almost unchanged with temperature between 350 and 455°C and was equal to about 0.3. G_2 also was larger in the oxygen-nitrogen mixture and increased rapidly with temperature. About 55% of the organic sulfur was released at 455°C. G_1 and G_2 both increased with holding time at 400°C in the oxidizing atmosphere, while f decreased as the holding time was increased.

CONCLUSIONS

Temperature is the most important factor influencing desulfurization in the three gases tested. The holding time at temperature is less important except in the case of hydrogen above 600°C where the sulfur content of the sample decreases with increased holding time. The experimental evidence indicated that this was due to the direct reaction of hydrogen with sulfur in the organic portion of the coal which takes place only at temperatures of 600°C or above. It was observed in the experiments that some inorganic sulfur was transformed into organic sulfur during the gaseous treatment. The fractions of inorganic and organic sulfur released as gases and the fraction of inorganic sulfur transformed into organic sulfur were estimated for each of the gas atmospheres and conditions of temperature and time used.

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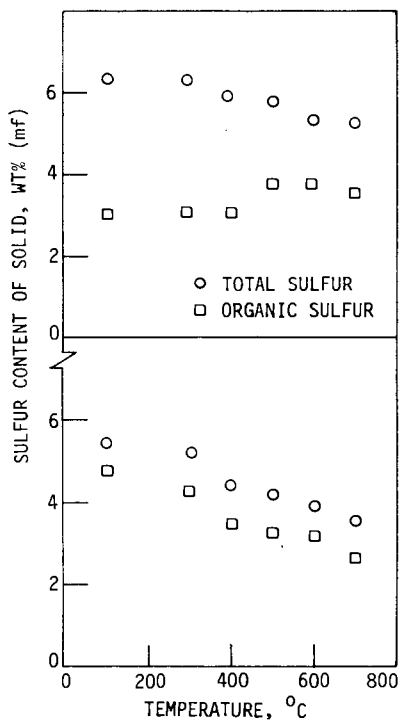


Fig. 1. Effect of temperature on the sulfur content of raw* and deashed coal heated in hydrogen.

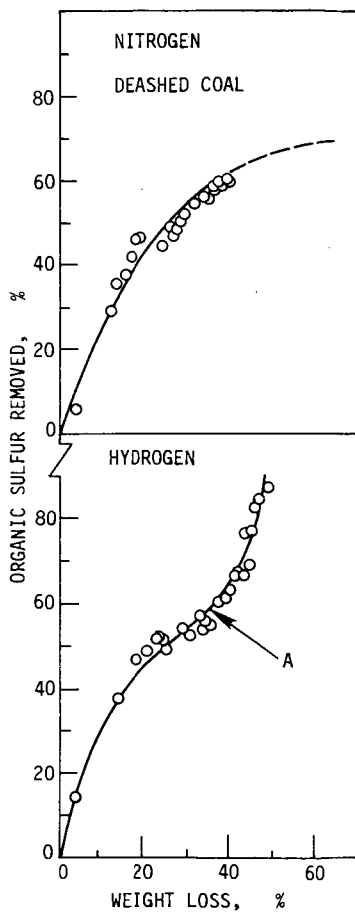


Fig. 2. Organic sulfur removal as a function of sample weight loss for deashed coal.

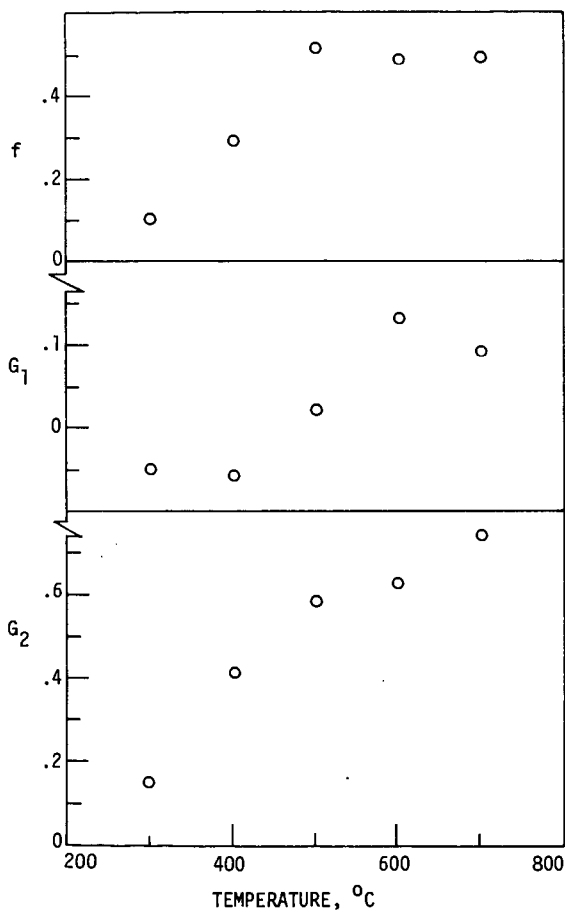


Fig. 3. Effect of temperature on the transformation of sulfur in a hydrogen atmosphere.